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**PROSPECTOR VII:
SMALL FUEL CELLS
FOR PORTABLE POWER
WORKSHOP**

October 31- November 3, 1994

Edited by

**Calvin R. Johnson & M. Frank Rose, Co-Directors
For the Prospector VII Board of Directors**

Sponsored by

**Space Power Institute
Auburn University, Alabama 36849
&
Army Research Office
Research Triangle Park, North Carolina 27709-2211**

**Held at the
Washington Duke Inn
Durham, North Carolina**

The views, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Defense position, policy, or decision, unless so designated by other documentation.

**M. Frank Rose, Director
Space Power Institute
Auburn University, Alabama**

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PREFACE

The Prospector Workshops, a series of highly focused workshops dealing with key issues associated with the science and technology of advanced power systems, had their origin in many conversations among outstanding technologists from all over the world. It became apparent that the difference between the state-of-the-art (SOA) and what these technologists saw for future needs was so large that new approaches to meet these needs was mandatory. As a result of this keen interest, a workshop format was conceived to bring the SOA and future needs closer together. The founders believed that a workshop's informal, yet intense review of advanced power systems' requirements, SOA, innovative ideas and concepts, technologies, and development programs, would create a new perspective of where certain critical power technologies were headed. There are a host of key issues which fall into this category of prime power, such as thermal management, advanced energy conversion, life support, automated systems and advanced diagnostic techniques, just to name a few. The sponsors believe that, due to the interdisciplinary nature of power systems, any new and successful approach will likely come from a group of technologists with diverse backgrounds rather than from those schooled in the "accepted" approaches. Thus, Prospector was created, in an attempt to identify and resolve some key issues regarding power-related systems.

The focus of preceding workshops of this Prospector series was on the production and management of electrical power and micro-electromechanical systems which might reduce the need for power or provide the techniques to more efficiently manage it. The focus of this workshop, Prospector VII, is on the options available to potential users in applications of small, portable fuel cell systems for man-portable military systems and commercial devices. The intent of this workshop is to incorporate the many advances in fuel cell design, power production techniques, chemistry and chemical engineering issues, fuels, and manufacturing opportunities into a concise strategy for significant advancements over the next decade.

As for the Prospector Workshop series, there is always something in a name. Just as the prospectors of old sometimes worked the tailings of old diggings searching for a missed nugget, we too reviewed the current research issues "looking for nuggets," before embarking on a search of new ground. For this we assembled a wide range of technologists -- engineers, physicists, manufacturing specialists, and managers representing the government laboratories, industry, and the university community. Three separate Working Groups were charged with evaluating the commercial and military applications, comparing and contrasting alternative fuel cell technologies, and defining and prioritizing the key research issues pacing fuel cell development.

In keeping with the tradition of the previous Prospectors, the workshop was patterned after the highly successful Gordon Conferences which have formal morning and evening sessions, leaving the afternoon free for recreation, small working group discussions or literature review at the participants' discretion.

The workshop was directed by a group of senior scientists from the Army Research Office and the Space Power Institute at Auburn University, a group which is familiar with the concepts of fuel cells and their many applications. The broad technical base represented by the Board of Directors resulted in a unique agenda which covered a cross-section of fuel cell technologies as well as the many opportunities which are presented to apply the fuel cell concept to military and commercial systems. Prospector VII's Board of Directors included Dr. Bob Guenther, Dr. John S. Kruger, and Dr. Richard Paur, (Army Research Office), Dr. Robert Hamlen (US Army Research Laboratory), and Mr. Cal Johnson and Dr. M. Frank Rose (Auburn University Space Power Institute).

The workshop organizers would like to express thanks to the staff of the Washington Duke Inn, Durham, North Carolina, and to the administrative staff of the Space Power Institute for organizing and managing the workshop. Special thanks are due to Ms. Jullie Campbell of the Washington Duke Inn and to Ms. Dana Latham of the Space Power Institute. Their efforts contributed greatly to the success of the workshop and to this archival record.

The pages that follow contain a detailed record of the workshop agenda, an Executive Summary, the results and recommendations of the Working Groups, copies of the individual technical presentations, and a list of the attendees, key technologists from government, industry and academia. We owe a special word of thanks to the attendees -- we sincerely appreciate your willingness to give your time and expertise to the discussions and technical presentations of this meeting. We sincerely hope that this document represents an accurate distillation of the workshop deliberations. It is, after all, your collective opinion which is archived here, and whatever impact this document has in the future is due to your thoughtful deliberations.

We hope to see many of you at Prospector VIII: Cutting Space Launch Costs, to be held March 21-24, 1995, in Park City, Utah.


M. Frank Rose, Co-Director
For the Board of Directors

EXECUTIVE SUMMARY

EXECUTIVE SUMMARY

This Workshop, Prospector VII, was held at the Washington Duke Inn, Durham, North Carolina, from October 31st through noon on November 3rd, 1994. The Workshop was held in response to a growing interest on the part of DOD and industrial sponsors in developing a means of assessing the current state-of-the-art in small fuel cells for portable power systems. This Workshop's three-day agenda included a series of background briefings, tutorials and technology updates, and concluded with several working group discussions which reached a consensus on three major topic areas. The event was sponsored by the Army Research Office and Auburn University' Space Power Institute, Auburn, Alabama. The findings, and details on how these were identified, are presented in the sections which follow.

Summary of Workshop Findings

A summary of the findings of the three Working Groups is stated below:

- Fuel cell state-of-the-art is approaching the Army's near-term mass/volume power system goals
- Fuel cells are a viable option to replace batteries in demanding, low power, mobile Army applications
- Best fuel cell candidates are pressurized hydrogen with PEM, chemical hydride with PEM, and direct methanol with PEM
- Mission-specific fuel cell designs are necessary to meet Army needs
- Key research issues include the selection and use of fuels and fuel storage methods, stack design and materials, electrolytes, catalysis, and systems engineering /innovative modeling techniques
- Range of Soldier System power systems could be filled by batteries, fuel cells, or a combination thereof, for a specific Army mission requirement
- Limiting attributes in fuel cell development are cost of manufacture, mass/weight challenges
- Technical issues could be resolved through R & D and modeling
- Military R & D will benefit by lower procurement costs from an increasing commercial fuel cell market in the range of 25 - 150 W
- Independent market survey is needed to identify dual-use cost share

Background of Prospector VII Discussions

The general topic, development of portable fuel cell power systems, evolved from ongoing ARO and ARPA small fuel cell programs and a prior Prospector Workshop which examined power sources that could be worn or carried by the individual soldier as part of the Soldier System concept. The three-day agenda included a series of background briefings, tutorials, and fuel cell technology updates. These briefings were designed to draw the participants' attention toward three major sets of criteria -- *requirements, capabilities, and program objectives which focus on small, man-portable fuel cell systems for military and non-military applications*. During the three days, participants examined a number of fuel cell technologies, comparing the currently favored PEM fuel cells (both hydrogen/air fuel and direct oxidation of methanol) with other types to determine whether there was reason to broaden the Army R&D program. Fuels were discussed in some detail, to document the positive and negative aspects of hydrogen and methanol as possible battlefield fuels, and to determine whether there was a case for other approaches (for example, the likelihood that very small reformers could be built for man-portable systems).

The focus of this meeting was on the options available to potential users in applications for man-portable military systems as well as industrial devices. The intent of the Workshop sponsor was to incorporate into a concise strategy fuel cell design, power production techniques, chemistry and chemical engineering issues, fuel types, availability, and production, and manufacturing/miniaturization techniques, so as to track fuel cell advancements over the next decade. The participants were divided among three Working Groups, namely (1) Civilian & Military Fuel Cell Applications for Commercial Products and Missions, (2) Alternative Fuel Cell Technologies -- Comparisons & Contrasts, and Key Research Issues Pacing Development -- Definitions & Priorities.

The Working Groups discussed the many tradeoffs in fuel cell selection criteria, various testing protocols, man-portable potential, performance measurement techniques, development issues, endurance trials, and other design options. At its conclusion, this Workshop attempted to clarify what might be done to influence specific improvements in fuel cell technologies for potential applications within the commercial sector and the military services with emphasis on man-portability and near-term, dual-use development opportunities.

Workshop Objectives

To guide participants, the sponsor developed the objectives listed below:

- Assess the state-of-the-art of fuel cells and fuel cell systems by characterizing innovative fuel cell conversion techniques and determining their applicability to DOD and non-military power systems;
- Identify the key research issues pacing development of (or limiting full development of) efficient, high power fuel cells/systems with acceptable lifetimes;

- Identify the major limiting factors which must be addressed as part of overall fuel cell/system design for man-portable applications;
- Prioritize and clearly define research issues, indicating the impact if successful;
- Provide research team milestones to track significant improvements in fuel cell technology over near-term and long-term development programs;
- Identify operational/environmental constraints such as materials, signatures, mass, and pollution which influence the applications or improvements envisioned; and
- Establish scaling laws, and wherever possible, use those laws to compare results with other methods of powering DOD and non-military systems.

Working Groups: Discussion Details

Group A: Applications for Commercial Products & Specific Missions

State-of-the-art fuel cell systems for low power (i.e., less than 1 kW) mobile Army applications were assessed and an H₂-air PEM reference system was defined. Hydrogen storage and re-supply logistics were discussed and system level heat transfer, scaling issues, and risks were addressed for the reference system. The state-of-the-art system is approaching the near-term mass and volume power system goals set by the Army.

It was concluded that fuel cells should be considered for use in low power mobile Army applications. Fuel cells should also be evaluated to replace existing battery systems and for use as battery chargers. The Working Group focused primarily upon 150 W system subject to the following requirements: long life (greater than 500 hrs), fast start/stop times, numerous start/stop cycles, freeze/thaw tolerance, rugged, soldier portable, attitude insensitive. Since each candidate application will have a different mix of operational and integration aspects, the Group found that specific power system designs would have to be created to accommodate specific mission requirements. As in other workshops on power system development, the participants pointed to the ultimate cost of fuel cell systems as the most important issue for commercial acceptance, probably established through a set of niche markets. Finally, several dual-use applications and potential commercial markets were suggested and discussed, and the Group concluded that insufficient information existed on the full market availability. A commercial customer base will emerge if a dependable, affordable fuel cell product becomes available. Thus, an independent market survey was strongly recommended.

Group B: Alternative Fuel Cell Related Technologies

The state-of-the-art fuel cell systems for low power (i.e., less than 1 kW) mobile Army applications were assessed in a general sense. The Group

concluded that the state-of-the-art (SOA) system is approaching the near term mass and volume power system goals set by the Army, and further technical progress appears to be readily achievable. *Only time and the necessary investment of R & D dollars seem to prevent the development of the Group's three prime candidates for near-term, small (portable) fuel cell applications.*

Working Group B focused on three power ranges, 10W, 150W, and 1000W systems by evaluating both fuel cell and non-fuel cell options. The participants recognized that some military and commercial applications could be technology-dependent, in that certain applications would employ only very specific types of technologies whenever mass and size were taken into consideration. Additional constraints placed upon the power system developer (by the user, or by outside agencies) could eliminate certain types of fuel cell systems completely, so care was taken not to immediately constrain discussions concerning possible applications and power system technologies. Key issues, such as operational and integration issues, as well as specific military/commercial applications, were left for other Working Groups to evaluate.

In its conclusion, the Alternative Fuel Cell Related Technologies Working Group stated that the over-riding positive and negative attributes which tend to limit small fuel cell development could be summed up in terms of cost of manufacture, and mass/weight challenges for the truly small, portable FC systems required by the Army. It was believed that technical issues, such as energy density, methanol cross-over, operating temperature limitations, optimal start-up characteristics and performance, and endurance (life cycle before replacement or refueling) would be solved or resolved through enhanced research, improved comprehensive modeling of systems, and better materials selection and optimization. Those issues were examined in detail by another Working Group.

This Group's final conclusion was that fuel cells should definitely be considered as a viable option for use in low power mobile Army applications, and an active Army R & D program was justified. As in discussions by others, this Group believes that fuel cells should also be evaluated to replace existing battery systems in selected demanding applications, and for use as battery chargers. *As a bottom line, the Group stated that, in the foreseeable future, the best candidates beyond lithium primary batteries were the following three fuel cell systems: pressurized hydrogen with PEM, chemical hydride with PEM, and direct methanol with PEM. As a non-fuel cell option, it was believed that metal-air batteries offered the best opportunity for development.*

Group C: Key Research Issues Pacing Development

The key research issues pacing development for low power fuel cell (FC) systems were identified and discussed in the Working Group C sessions. The Group concluded that there were a number of research issues which could be arranged into five categories. These categories related to FC sub-systems, techniques and methods for improvements in design, safety aspects, or a better application of materials and fuel cell-related chemistry. Also, the Group determined that the most relevant key research

issues, distributed into five major categories, were the selection and use of fuels and fuel storage methods, stack design and materials, electrolytes, catalysis, and systems engineering /innovative modeling techniques for fuel cells.

The Group limited itself to discussing portable power requirements which are necessary for the individual soldier. The requirements were divided into two primary functional areas. First, microclimate cooling which requires power on the order of 150 watts and up; and second, all other electronic devices requiring power from 5 watts up to ~50 watts. As shown in Figure 1, potential packaging for power requirements have been divided into these two broad functional areas, microclimate cooling and others.

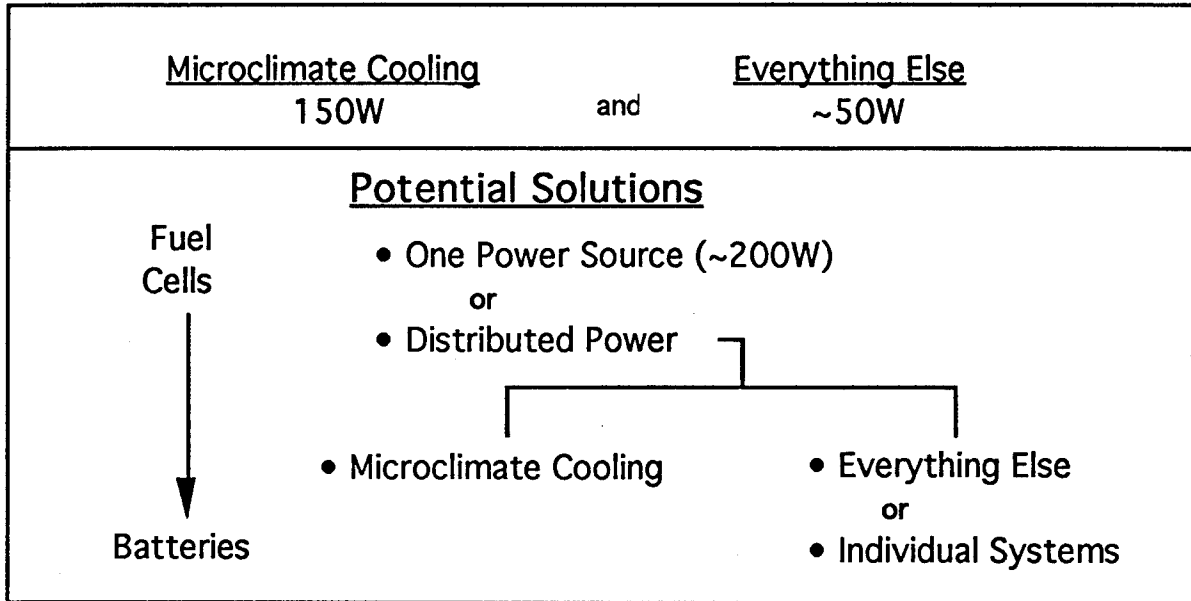


Figure 1. Individual Soldier Power Requirements and Potential Solutions

In one category a single power source (approximately 200 watts) will provide all the currently identified energy needs for the individual soldier. Alternatively, a distributed power system of two major types could be developed. One power system would be only for microclimate cooling, the second power system would include everything else. Furthermore, this second power system can be further divided into two modes; a single power unit that supplies all the needs, or distributed individual systems for the various components. *A single source 200 watt unit would, by necessity, probably be a fuel cell system.*

On the other hand, as further depicted in Figure 1, sub-dividing the power source into a distributed system of various sizes could result in some systems capable of being handled by batteries. So the range of power needs could be fulfilled by fuel cells or batteries, depending on the specific mission. As shown in Figure 2, it is envisioned that a 50 watt power system will be met by two devices of 25 watts each. The energy requirement is estimated to be ~300 watt hours requiring less than 1 kg mass and 1 liter of volume. *This energy need possibly could be met by fuel cells or by batteries.*

<u>Power(W)</u>	<u>Energy(W-Hr)</u>	<u>Wt(Kg)/V(L)</u>	<u>Technology</u>
25	300	1/1	Battery/Fuel Cell
150	>600 - 10,000	5/4	Fuel Cell
>150			
<u>Potential Fuel Cell Solutions</u>			
<ul style="list-style-type: none"> • He/PEM • MeOH/PEM • MeOH/SOFC 			

Figure 2. Power/Energy Targets for Future Work and Potential Solutions

On the other hand, the 150 watts and above power source will require energy levels on the order of 600 up to ~10,000 watt-hrs. It will need to weigh less than 5 kg and have a volume less than 4 liters. *This particular need will certainly require a fuel cell.* The working group decided that the fuel cell technologies which are available at this time and are most amenable for these applications include the hydrogen PEM, the methanol PEM and the methanol solid oxide fuel cell (SOFC). As a result, all research issues discussed revolved around those three FC systems.

The Group concurred with the Sponsor's Charge, in that the system's energy density is of paramount importance in establishing parameters, planning factors, limitations, markets, and options for technology development. Furthermore, in evaluating the "small system," the definition for "small" should be defined as "any system less than 1kW at the upper end."

Finally, the Working Group acknowledged that the development of fuel cell systems of 25 watts up to less than 150 watts for military applications could lead to numerous applications in the commercial sector. Although military applications were the primary motivator for this particular workshop, one should consider the commercial spin-offs due to the vastly increased market and investment opportunity. Consequently, it is the opinion of the Group that an emphasis on research to resolve these fuel cell technical issues will benefit civilian markets as well as military needs, and this "dual market opportunity" should be pursued.

Summary

In summary, participation in the Prospector VII Workshop offered a unique opportunity for scientists and engineers from industry, universities, and

government to jointly develop a strategy to advance the technology for portable fuel cell systems for military and civil applications. On behalf of all DoD and industrial users who will benefit from this report, the members of the Board of Directors wish to thank each participant for providing detailed, constructive, and candid comments on a development strategy for portable fuel cell power systems. Thank you for a job well done!



Frank Rose

Co-Director

Board of Directors - Prospector VII

Dr. Richard Paur, Dr. Bob Guenther, Dr. Jack Kruger; US Army Research Office
Dr. Robert Hamlen, US Army Research Laboratory
Dr. M. Frank Rose, Mr. Cal Johnson; Auburn University's Space Power Institute

FINAL AGENDA
Prospector VII: Small Fuel Cells for Portable Power

Washington Duke Inn
Durham, North Carolina
October 31 - November 3, 1994

Monday Evening Registration Period (October 31)

- I. Registration (Foyer) & Reception in Washington Duke Inn Salon A B (6-7:00pm)
- II. Welcome and Overview (Salon AB) - Board of Directors, Sponsors (7:00-7:15pm)
- III. Plenary Session (20-25 minutes each, 7:15-9:30pm) - Mr. Cal Johnson, Auburn (Chairman)
 - .. "ARO Interest in Portable Fuel Cells," Dr. Dick Paur (ARO)
 - .. "Natick's Fuel Cell Requirements for the Soldier System," Mr. Brad Laprise (Natick)
 - .. "Silent Energy Sources for Tactical Applications (SIESTA)," Mr. Walter Taschek (Belvoir)
 - .. "Competing Technologies for Power Generation," Dr. Frank Rose (Auburn's SPI)
 - .. "High Performance Fuel Cell Initiatives at ARPA," Dr. Lawrence Dubois (ARPA)

Tuesday Morning (November 1)

- I. Continental Breakfast: Salon AB (7:30-8 am)
- II. Introductory Comments: Technology Update - Dr. Frank Rose, Auburn (8-8:15am)
- III. Technology Update Session 1 (15 minutes each, 8:15-10:00 am) Dr. Frank Rose, Chairman
 - .. "Status of ARL Battery Programs," Dr. Bob Hamlen (ARL)
 - .. "Proton Exchange Membrane Fuel Cells - Basic Research to Technology Development," Dr. Supramaniam Srinivasan (Texas A&M)
 - .. "Fuel Cell Development Program for the Army," Mr. Dave Bloomfield (Analytic Pwr)
 - .. "Overview of PEM Fuel Cell Research at ARL," Dr. Michael Binder (ARL)
 - .. "Al/Air Semi-Fuel Cells for Portable Power Applications," Dr. Bhaskara Rao (Explores)
- IV. Coffee Break/Displays & Posters in Lobby Area (10:00-10:30am)
- V. Technology Update Session 2 (15 minutes each, 10:30-12 noon) - Dr. Bob Guenther, ARO
 - .. "System Integration Aspects of Small Portable Devices," Dr. Scott Hynek (Arthur D. Little)
 - .. "Energy Storage: Manpack Hydrogen Tanks," Dr. Ira Kuhn (Directed Technologies)
 - .. "Oxygen Storage Systems," Mr. Jerry Chodil (Ball Aerospace)
 - .. "Direct Methanol PEM Fuel Cell Development," Dr. Bob Savinell (Case Western Res)
 - .. "Evaluation & Modeling of PEM Fuel Cells," Dr. Ron Mann (Royal Military Col Canada)
 - .. "Electrodeposition of Nafion on Electrode Structures," Dr. Bruce Tatarchuk (Auburn)
 - .. "Innovative Laminar Structure Concept," Dr. Eugene Smotkin (Illinois Inst of Tech)
- VI. Poster Session/Demo Period (Some individuals have expressed interest) (12-1pm)
- VII. Lunch (No Host)
- VIII. Dinner at Washington Duke Inn, Hosted by Auburn University's Space Power Institute. Center Room: Menu - "Ambassador Buffet," Salad, Fruit, Vegetables, Assorted Desserts (6-7:30pm)

Tuesday Evening

- IX. Technology Update Session 3 (15 minutes each, 7:30-9:30pm) - Dr Bob Hamlen, USARL
 - .. "Low Temp Solid Oxide Fuel Cells at Argonne," Dr. Mike Krumpelt (Argonne Lab)
 - .. "Heat Transfer and Cooling Challenges in Fuel Cell Development," Dave Bloomfield (Analytic Power)
 - .. "Field Production of H₂: Wet or Dry Manufacture from Military Transport Fuels," Dr. Bill Peters (MIT)
 - .. "Update on Battery Workshop (Prospector III)," Dr. Bob Guenther (ARO)

a/o 10/26/94; 11am

Wednesday (November 3) - Dr. Dick Paur (ARO), Chairman

- I. Charge to the Working Groups - Dr. Dick Paur
- II. Continental Breakfast: Salon A (7:30-8 am)
- III. Working Group Deliberations (Salons A, B & C; 8am - 12 noon)
- IV. Lunch (No Host)
- V. Working Group Deliberations (Salons A, B & C; 1:30-5:30 pm) (Tentative)
- VI. Dinner (No Host)
- VII. Working Group Deliberations, Briefing Preparation (Salons A, B & C; 7:00-10:00 pm)

Thursday (November 4) - Dr. Jack Kruger (ARO), Chairman

- I. Report of Working Groups, Plenary Session (Salon AB; 8:00-11:30pm)

Note: Workshop Message Center is located in the hall area outside Salon B.

a/o 10/20/94; 4pm

INTRODUCTION

INTRODUCTION

Prospector VII is the seventh in a series of workshops dealing directly with advanced technologies applied to the individual soldier's needs. Other workshops investigated the alternative power sources (batteries, capacitors, radioisotope-based generators, etc.) for the Soldier System concept, and a fuel cell power system was the only remaining form not studied on an individual basis as a result of recommendations made by participants in prior workshops. This Workshop completes the remaining recommendations and underscores the growing interest in, and need for, small, portable fuel cell systems on the part of DOD and industrial sponsors.

As background for this topic, in November of 1990, the first workshop on Mobile Tactical Battlefield Power Technology was held at the request of the Army Research Office. One of the major findings was the identification of a need for research and development to improve Army power technology at the low end of the scale. Power technology, up to 500 W, and man portable, is considered absolutely necessary to assure the effectiveness of the Army's mobility. Furthermore, in years past, such issues as autonomous operation time, reliability, scaling, and cost had not been clearly defined for power systems. These, and other such unresolved issues in recent years, emphasized the need for separate workshops dedicated to sub-sets of the Army's power needs. Subsequent workshops, carefully focused, were conducted to address those particular sub-sets. As the Prospector series evolved, workshop topics of particular interest to the Army were studied, to include electrochemical power sources, small engine-generator power systems, electric actuation and its applications, radioisotope thermoelectric generators, the new and emerging technology of micromachines, the thermal management aspects of power systems, and the most current topic, small, portable fuel cell power systems for the individual soldier.

The **Workshop goals**, as determined by the Board of Directors, were to:

- Assess the state-of-the-art of fuel cells and fuel cell systems by characterizing innovative fuel cell energy conversion techniques and determining their applicability to DOD and civil power systems;
- Identify the key research issues pacing the development of (or limiting full development of) efficient, high power fuel cells/systems with acceptable lifetimes;
- Identify the major limiting factors which must be addressed as part of overall fuel cell/system design for man-portable applications;
- Prioritize and clearly define research issues, indicating the impact if successful;
- Provide research team milestones to assure significant improvements in fuel cell technology over near-term and long-term development programs;

- Identify operational/environmental constraints such as materials, signatures, mass, and pollution which influence the applications or improvements envisioned; and

- Establish scaling laws, and wherever possible, use those laws to compare results with other methods of powering DOD and civil systems.

To accomplish these goals, a group of distinguished scientists and engineers, active in the field, were invited to present, in several overview talks, current perceptions of the state-of-the-art in fuel cell power systems. Each speaker was to address key issues, operational and environmental constraints, and potential military and commercial applications for small, portable fuel cell systems from their own particular vantage points in DOD agencies, academic institutions, and research laboratories. These speakers constituted the plenary session, and the agenda for that session is shown in Table 1.

SMALL, PORTABLE FUEL CELL SYSTEMS PLENARY SESSION

Chairman: Mr. Calvin Johnson (AU)

- "ARO Interest in Portable Fuel Cells," Dr. Dick Paur (ARO)
- "Natick's Fuel Cell Requirements for the Soldier Sys," Mr. Brad Lapris, Natick
- "Silent Energy Sources for Tactical Applications," Mr. Walter Taschek, Belvoir
- "Competing Technologies for Power Generation," Dr. Frank Rose, Auburn Univ
- "High Performance Fuel Cell Initiatives at ARPA," Dr. Lawrence Dubois, ARPA

Table 1.

As confirmed by the Plenary speakers, progress, and interest in, small, portable fuel cells is increasing steadily. The military is looking for smaller fuel cells with increased energy density, relative ease of use (little training required), lower cost per unit energy, increased power density, and reduced disposal problems. For obvious reasons, special features, such as a "remaining energy indicator (REI)" are desirable. What is the state-of-the-art? Successes in ARPA's direct methanol fuel cell are evident. That technology is approaching power densities of approximately 0.2 W/cm^2 . In another area, the Army Research Office is lowering cost and improving performance of the 150 - 250 W hydrogen units. One Plenary speaker reminded the audience that operation of PEM and SOFC fuel cell stacks on H_2 /air mixtures at greater than 1 kW/kg and 1 kW/L should be possible. Natick's Gen II (second generation Soldier System) prototype device was briefed to the participants. Another speaker detailed the power requirements

and what each power milestone represented in terms of a soldier's improved combat capability when fuel cell options were applied. A last speaker talked to novel thermophotovoltaic generator systems with tuned line emitters as a new technology which could be competitive for applications as small, portable power systems. *The Plenary Session's themes rapidly pointed to the advantages of refuelable power sources which are attitude-insensitive, silent, possess a low thermal signature, are safe and easy to operate, and are relatively cost-effective. A hybrid system or some novel approach would not be ruled out, nor would multiple types of power sources be discounted due to varying military mission scenarios, both in duration and peak energy demands.*

To assist the workshop process after the overview talks, other speakers concentrated on specific portions of FC development, and the technologies which are involved in each. Another group of fuel cell scientists and engineers presented technology summaries describing the state-of-the-art, assessing the near-term state-of-the-art, and giving their personal opinions of the ultimate limits with some considerations for practicality. The Agenda for Technology Sessions I, II & III is shown in Table 2.

The remainder of the Workshop was spent in smaller Working Groups where discussions centered around the following three main topic areas:

- **Civilian and Military Fuel Cell Applications** for Commercial Products and Specific Missions
- **Alternative Fuel Cell Technologies**
- **Key Research Issues** Pacing Development - Definitions & Priorities

Charge to Participants. As stated earlier, the individual Working Groups were charged with finding the best development paths and technology solutions for small, portable FC systems. "Small" was defined by the sponsor as less than 1 kW at the upper end and as small as practical at the lower end. *The sponsor emphasized that the complete power system had to be carried, so the energy density of complete systems was what really counted.* Additionally, Group I (Applications) was to assess the SOA, assess whether it was feasible to go to smaller units to attempt to replace batteries, assess commercial markets and issues, and address system issues of stack integration, attitude sensitivity, shelf life, start-up time, disposal, and safety. Group II (Alternatives) was to identify alternatives to FC in small size ranges, assess alternative FC technologies for systems under 1 kW, summarize advantages and disadvantages of alternate technologies when compared to fuel cells, and identify self-contained options and size range of application (assess the marketability of fuel cells) in military and commercial markets. Group III (Research Issues) was charged with developing a consensus of the leading technical "show-stoppers" limiting the rapid development of small FC units, prioritizing these leading technologies, identifying SOA performance and goals for future work, summarizing the potential strategies for near-term improvements and "breakthroughs," and identifying the basic scientific support needs for FC improvements.

With this set of questions and challenges, Working Group members were to seek out new ideas, identify the technological impediments, and secure comparisons of traditional and non-traditional technologies. The summaries of those discussions, and the respective Group briefings to the entire body of participants, are included in these Proceedings.

TECHNOLOGY UPDATE SESSION I Chairman: Dr. Frank Rose (SPI)

- "Status of ARL Battery Programs," Dr. Bob Hamlen, Army Research Lab
- "Proton Exchange Membrane Fuel Cells - Basic Research to Technology Development," Dr. Supramaniam Srinivasan, Texas A&M
- "Fuel Cell Development Program for the Army," Mr. Dave Bloomfield, Analytic Power
- "Overview of PEM Fuel Cell Research at ARL," Dr. Michael Binder, Army Research Lab
- "Aluminum/Air Semi-Fuel Cells for Portable Power Applications," Dr. Bhaskara Rao, Explorex

TECHNOLOGY UPDATE SESSION II Chairman: Dr. Bob Guenther, ARO

- "System Integration Aspects of Small Portable Devices," Dr. Scott Hynek, Arthur D. Little
- "Energy Storage: Manpack Hydrogen Tanks," Mr. Ira Kuhn, Directed Technologies, Inc
- "Oxygen Storage Systems," Mr. Jerry Chodil, Ball Aerospace
- "Direct Methanol PEM Fuel Cell Development," Dr. Bob Savinell, Case Western Reserve University
- "Evaluation & Modeling of PEM Fuel Cells," Dr. Ron Mann, Royal Military College of Canada
- "Electrodeposition of Nafion on Electrode Structures," Dr. Bruce Tatarchuk, Auburn University
- "Innovative Laminar Structure Concept," Dr. Eugene Smotkin, Illinois Institute of Technology

Table 2.

TECHNOLOGY UPDATE SESSION III

Chairman: Dr. Bob Hamlen, ARL

- "Low Temp Solid Oxide Fuel Cells at Argonne," Dr. Mike Krumpelt, Argonne Laboratory
- "Heat Transfer and Cooling Challenges in Fuel Cell Development," Mr. Dave Bloomfield, Analytic Power, Inc
- "Field Production of H₂: Wet or Dry Manufacture from Military Transport Fuels," Dr. Bill Peters, MIT
- "Update on Battery Workshop (Prospector III)," Dr. Bob Guenther, ARO

Table 2 (Continued).

In a final general session, each Working Group Chairman presented a summary of his respective group's deliberations and findings to a final Plenary Session. As expected, there was considerable discussion of the separate findings, as well as a general consensus on a number of key points arrived at in separate session. These findings have been incorporated as accurately as possible into the Executive Summary and the individual Working Group Summaries.

Three events, a demonstration, display, and poster session of portable fuel cell technology, provided enrichment to the technical presentations (Table 3).

- A. Demonstration/Display: Analytic Power's Operational Fuel Cell (Bloomfield)
- B. Display: CECOM's Fuel Cell Development Device (Taschek)
- C. Poster Session: Factors Governing On-Line Hydrogen Generation from Aluminum in Alkaline Solution (Rao)
- D. Poster Session: PEM Fuel Cells - Basic Research to Tech Development (Srinivasan)

Table 3.

The fifty-three participants, drawn from industry (19), academia (12), and government agencies and laboratories (22), represent a balanced cross-section of fuel cell researchers, users, and technologists.

The remainder of this document is a collection of the Workshop presentations and summaries from the individual Working Groups. The Appendices contain additional documentation or background papers as necessary to provide a complete written record of the events of Prospector VII.

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PLENARY SESSION

PLENARY SESSION

Chairman: Mr. Calvin Johnson (AUSPI)

The Plenary Session began with a welcome by Dr. M. Frank Rose and Dr. Dick Paur on behalf of the Workshop's Board of Directors. Their comments to the group constituted a reiteration of the Workshop objectives and a reminder from Dr. Paur that he would be providing some detailed guidance to each Working Group just prior to the "charge to the Workshop body." Both presentations would be presented following the last Technology Update Session on Tuesday evening, November 1st.

The Plenary Session speakers confirmed that progress, and interest in, small, portable fuel cells is increasing steadily. The military is looking for smaller fuel cells with increased energy density, relative ease of use (little training required), lower cost per unit energy, increased power density, and reduced disposal problems. For obvious reasons, special features, such as a "remaining energy indicator (REI)" are desirable. What is the state-of-the-art? Successes in ARPA's direct methanol fuel cell are evident. That technology is approaching power densities of approximately 0.2 W/cm².

In another area, the Army Research Office is lowering cost and improving performance of the 150 - 250 W hydrogen units. One speaker reminded the audience that operation of PEM and SOFC fuel cell stacks on H₂/air mixtures at greater than 1 kW/kg and 1 kW/L is possible. Natick's Gen II (second generation Soldier System) prototype device was described in terms of its major program elements and five major subsystems. Of the five subsystems, only the Protective Subsystem does not require power.

Another speaker detailed the power requirements and what each power milestone represented in terms of a soldier's improved combat capability when fuel cell options were applied. A last speaker talked to novel thermophotovoltaic generator systems with tuned line emitters as a new technology which could be competitive for applications as small, portable power systems. *The Plenary Session's themes rapidly pointed to the advantages of refuelable power sources which are attitude-insensitive, silent, possess a low thermal signature, are safe and easy to operate, and are relatively cost-effective. A hybrid system or some novel approach would not be ruled out, nor would multiple types of power sources be discounted due to varying military mission scenarios, both in duration and peak energy demands.*

The Agenda for this session was as follows:

- "ARO Interest in Portable Fuel Cells," Dr. Dick Paur (ARO)
- "Natick's Fuel Cell Requirements for the Soldier System," Mr. Brad Lapris, U. S. Army Natick RD & E Center
- "Silent Energy Sources for Tactical Applications," Mr. Walter Taschek, Belvoir

- "Competing Technologies for Power Generation," Dr. Frank Rose, Auburn Univ
- "High Performance Fuel Cell Initiatives at ARPA," Dr. Lawrence Dubois, ARPA

Speakers' Summaries - Enclosed at Front of Each Presentation

Each speaker provided a 1-2 page narrative/summary, with a copy of the lecture materials (overhead projection viewgraphs). **The remainder of this section is a compilation of the Plenary Session lectures and their summaries.** They are reproduced here as submitted by the Workshop organizers and speakers.

PERSON-PORTABLE ELECTRIC POWER

October 31 - November 3, 1994

Dr. Richard J. Paur

Electrochemistry
Chemical & Biological Sciences Division
Army Research Office

The Army Research Office's Battlefield Power Program has a definite Soldier System focus. That focus is represented by the numerous technology options and the route(s) taken by ARO to bring these competing options to closure culminating in a well-defined Soldier System power source. Some interesting conclusions have evolved during the past four years as a result of six workshops, SBIR, HUB, and URI programs, and several assessments of the state-of-the-art of technologies which could meet the requirements of small, compact power supplies for the individual soldier. Figure 1 describes the route(s) used to evaluate power source options over the past four years. Specific conclusions reached along the path are as follows:

a. Mobile Tactical Battlefield Workshop: Largest payoff from concerted R&D efforts is in the smaller power source applications, and politics neglected, small nuclear power units would be truly enabling. Issues which evolved led to further study on such issues as ways to use standard military fuels for hydrogen production for fuel cells, electrode structure development to capitalize upon number of cycles and depth of discharge of power storage devices, advanced thermal management and heat exchanges techniques, and methods of hydrogen storage. Fuel cells appear to be the most desirable, longer term solution, and a future workshop should study this option, as well as batteries, in more detail.

b. Radioisotope Power Workshop: This concept had desirable attributes to include an enormous energy density, adaptability to a wide range of power densities, isotopes which were suitable for power applications were byproducts of nuclear reactor operations, technology is highly developed, and offers a wide range of options for energy conversion. On the less desirable side, this technology has a power source which cannot be turned off, requires a massive shield for fuels, portends some environmental and health issues in manufacture and use, is expensive, and is probably publicly unacceptable.

c. Advanced Battery Workshop: Although most SS missions can be handled by battery technology, continued concerns on environmental issues and safety, highly influenced by civil and industrial applications, will impact Army use. Lithium chemistry is the basis for the most promising battery technology for the SS. Fuel cells and small rotating machinery can meet the power and mass requirements for the most demanding SS currently defined (5/92). As a bottom line, power technology is underfunded.

d. Small Engines for the SS: General conclusions were that there is enormous potential for improving small engine performance through the innovative use of materials and technologies. OSHA regulations will become restrictive, hence environmental and safety concerns will play a greater role in Army power technology. The Army is investing in the appropriate small engine power technologies, although power lags device and electronics development. Beamed microwave radiation was deemed to be unmanageable. As a bottom line, rotating machines could be ready in the near-term but has significant signature problems.

e. Micromachine and the SS Program: The opportunity is there for micromachines to be placed into sensors in support of the SS, but the micromachine technology does not seem to be as directly applicable to power generation. As shown in the figure, micromachines enter the SS design later.

Conclusions: General conclusions were that rotating machinery and fuel cells could support higher energy applications such as microclimate cooling. Rotating machinery had significant signature problems. MEMS was seen to be best suited to sensor applications. Most Soldier System applications require only a few watts of power and can be handled by batteries. Fuel cells appear to be the most desirable, longer term solution.

Recent Accomplishments/Milestones: The ARPA direct methanol fuel cell program demonstrated that multicell DMFC's with power densities approaching 0.2 W/cm^2 can be made. The SS community has stated unequivocally that they do not want to "wear" engine generator power systems. The ARO 150-250 W fuel cell program has lowered cost and improved the performance of small hydrogen systems. Most of the program has been transferred to CECOM-Belvoir. Both ARL and CECOM now have active fuel cell programs targeted at small FC's. SS designers are more interested than ever in better small power systems. "Better" is defined as increased energy density and power density, less expensive per unit energy, easy to use (little training involved), possesses a remaining energy indicator (REI), safe and reliable, and less disposal problems.

As an introduction to the Group discussions, each Working Group is being provided with a list of questions and thoughts which it should consider while evaluating its particular portion of the Workshop agenda. The Applications Group should address scaling laws for fuel cells, water management, start-up time requirements, and commercial applications for small fuel cells. The Alternative Technologies Group should address the relative development status of non-fuel cell technologies over the past several years, optional technologies to hydrogen and methanol for fuel cell power production, competitive power sources to fuel cells for this particular application, and the time frame until selected technologies would be ready for use in the field. The Key Research Issues Group needs to assess the state-of-the-art of fuel cell components and systems, prioritize a list of show-stoppers, and assess the value of more theoretical approaches to research issues.

Person-portable Electric Power

Richard J Paur

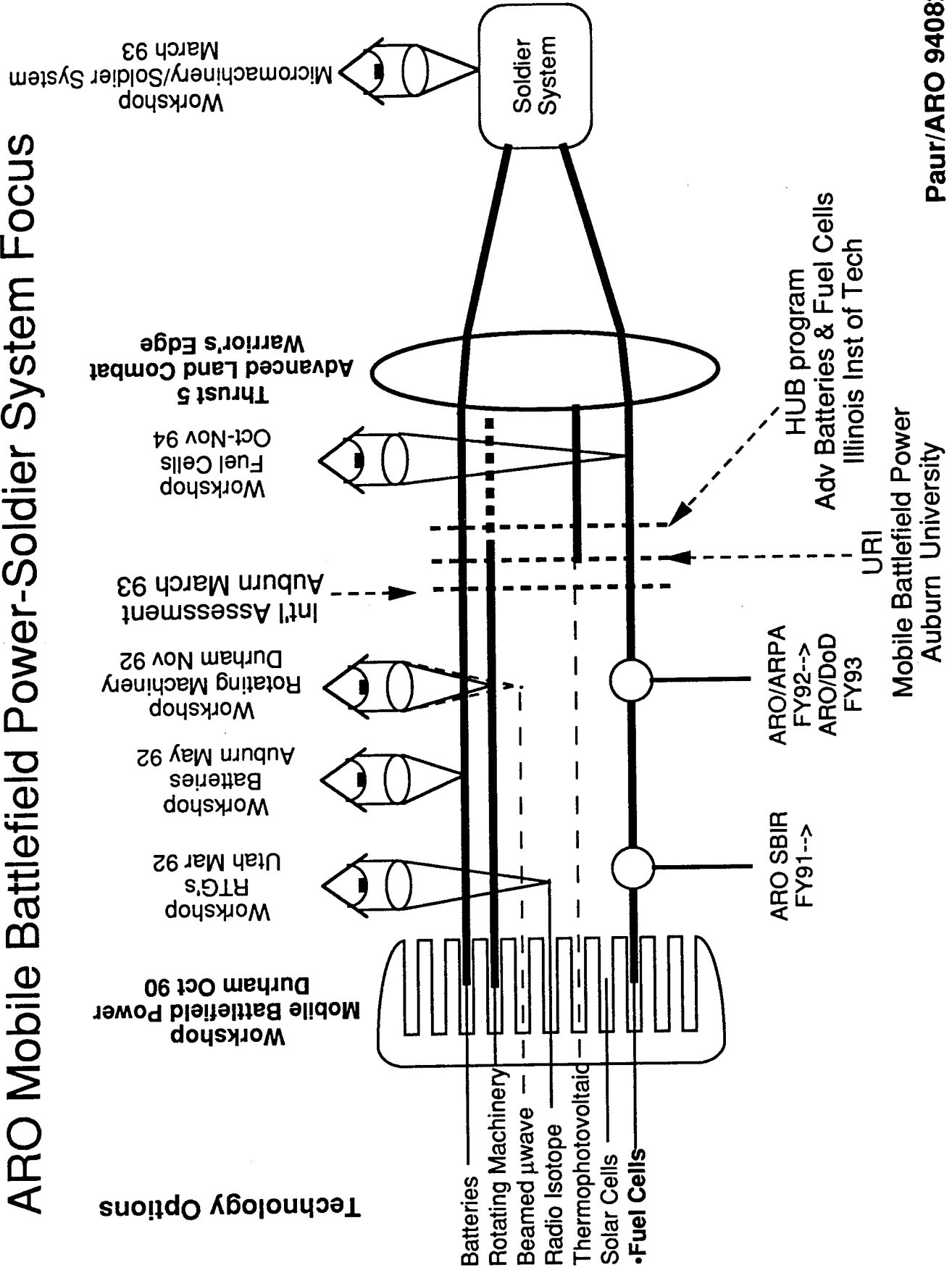
Chemical & Biological Sciences
Division

Electrochemistry

Presented at Prospector VII:
Small Fuel Cells for Portable Power
Washington Duke Inn
Durham, North Carolina
31 Oct -- 3 Nov, 1994



ARO Mobile Battlefield Power-Soldier System Focus



Mobile Tactical Battlefield Power

30 Oct - 1 Nov 1990

Most Interesting Conclusions

- Largest payoff from concerted R&D efforts is in the smaller power source applications
- Neglecting political and environmental concerns, small nuclear power units in the battlefield would truly enabling, extending autonomy time by orders of magnitude
- Issues
 - Hydrogen storage
 - Standard fuel decomposition for hydrogen production
 - Improvement of thermoelectric energy conversion
 - Advanced thermal management and heat exchange
 - Electrode structures to increase both the number of cycles and depth of discharge
 - Application of nuclear technology optimized to Army needs

Radioisotope Power Technologies and Applications

22-25 Mar 1992

Desirable attributes:

Enormous energy density

Systems can be made with a wide range of power densities
Isotopes suitable for power applications are byproduct of nuclear reactor operations

Isotope power systems are highly developed and reliable

Wide range of options for energy conversion

Less desirable attributes:

Isotope power cannot be turned on and off

Massive shield in required for some isotope fuels

Environmental/health issues associated with both manufacture and use of isotope systems

Nuclear powered systems in general have a dismal public image

Most desirable isotopes are expensive

Radioisotope Power Technologies and Applications

22-25 Mar 1992

Summary Conclusions:

- Many potential terrestrial applications for RTG technology are not currently being pursued**
- Public perception, cost of materials, and environmental/safety issues have tended to limit application to niche technologies**
- Factors of 2-5 improvement in efficiency of conversion is near term state-of-the-art**
- The isotope manufacturing industry in this country is in poor shape, border on collapse, especially for applications using large quantities**
- Before widespread applications can be achieved, public perception of nuclear systems must significantly change**

Advanced Battery R&D

27-28 May 1992

Most Interesting Conclusions

- Most Soldier System missions can be handled by battery technology, however, there will be continued safety/environmental concerns which will impact Army uses.**
- Civil and industrial applications are highly influencing battery technology and strongly influence what is available for military applications.**
- Lithium chemistry is the basis for the most promising battery technology for the Soldier System.**
- Fuel cells and small rotating machinery can meet the power and mass requirements for the most demanding Soldier System currently defined. Battery technology is marginal, and, for the most demanding mission, requires major R&D.**

Small Engines for the Soldier System

9-11 Nov 1992

General Conclusions

- There is enormous potential for improving small engine performance through the innovative use of new materials and technologies.
- OSHA regulations will become more restrictive - hence environmental and safety concerns will become an increasingly more important factor in Army power technology.
- Army is investing in appropriate small engine power technology
- Power technology generally lags electronics and device technology development
- Power technology is underfunded
- Beamed μ wave radiation was judged to be unmanageable

General conclusions from the series of workshops

- **Battery technology could handle most applications related to the Soldier System**
- **Rotating machinery and fuel cells could support higher energy applications such as microclimate cooling.**
- **Rotating machinery could be ready near term but had significant signature problems; fuel cells appeared to be the most desirable, longer term solution.**
- **MEMS was seen to have more application to sensors than to power generation.**

What has happened in the last two years?

- The ARPA direct methanol fuel cell program was successful in demonstrating that multicell DMFC's with power densities approaching 0.2 watts/cm² can be made.
- The Soldier community has stated unequivocally that they do not want to 'wear' engine-generator power systems (this is largely due to their perception of the signature, and also to some extent due to physiological concerns).
- The ARO program in 150-250 watt fuel cells has made progress in lowering the cost and improving the performance of small hydrogen systems. Most of the program has been transferred to CECOM-Belvoir. Both ARL and CECOM now have active fuel cell programs targeted at small fuel cells.
- The Soldier System designers are more interested than ever in better small power systems.

Basic assumption is that the military is 'always' going to be looking for better power sources - current emphasis is on smaller units

Better means:

- Increased energy density**
- Easy to use - little training involved**
- Less expensive / unit energy**
- Increased power density**
- Remaining energy indicator**
- Safe and reliable**
- Lessened disposal problems**

Working group 1 - applications

One of the short term needs facing designers working on soldier modernization programs is the need to reduce the number of batteries and improve the energy density of small power sources. Laprise, Taschek, Guenther

- How well do we understand scaling laws for fuel cells down to the 20 watt range? Will the system's ancillary hardware (eg, fuel tanks) become a relatively large part of the system and lower the energy density below that of batteries? What are the benefits of high pressure systems if the total watt-hours required are small?
- Are small units going to be able to tolerate being turned on and off - water management? What kind of startup times are envisioned? For microclimate cooling a startup of a few minutes may be OK, but for radios a much quicker response is required.

Working group 1 - applications (cont)

- **What commercial applications might use small fuel cells?
The Army ordered a thirty day supply of high performance batteries shortly before the end of Desert Storm; this supply turned into a several year peace-time supply of batteries; suppliers need to have a more or less constant demand for their product if the price is to be reasonable.**

Working group 2 - alternative technologies

At least two kinds of questions: Have any other (non-fuel cell technologies) made significant progress in the last few years; Are there any fuel cell technologies besides hydrogen and methanol PEM systems that are likely to deliver high energy density systems in power ranges below a kilowatt in the next decade or so? Rao, Rose

- Hydrogen vs methanol? This is the first workshop in this ARO series where it is true that there is a promising direct methanol fuel cell.
- Assess the state-of-the-art in hydrogen and direct methanol fuel cells. What is the likelihood of having systems available for field use in 2 years? 5 years? 10 years? The system must be able to operate in a battlefield environment, over a wide temperature range, and be attitude insensitive.

Working group 2 - alternative technologies (cont)

- What alternative technologies are the strongest competitors for fuel cells in the 10 to 50 watt range, the 150-250 watt range, the 350-1000 watt range? How do the choices depend on mission length? (short missions are < 5 hours, intermediate missions are 8-12 hours, and long missions are > 12 hours.)

Working Group 3 - key research areas

Research is currently moving forward in many areas

- **Hydrogen systems**
 - Fuel delivery - Bloomfield, Peters
 - Water/thermal management
 - Membranes - should we expect new materials?
 - Oxidants - is there a reasonable alternative to ambient air?
At what watt-hour range can you 'bottle' the oxidant?

- **Methanol systems**
 - Catalysts - what basis is there to expect significant advances
 - Membranes - several promising approaches - Savinell, Smotkin
 - System integration issues - ADL, JPL

Working Group 3 - key research areas (cont)

- Assess the state-of-the-art of fuel cell components - which areas can be expected to make reasonable progress under the current level of funding? Which areas would truly benefit from modest increases in funding vs areas where time (and developments in other areas of chemistry, materials science or engineering) appears to be more important than funding?
- Prioritize any likely 'show-stoppers' or difficult problems which may not yield in the foreseeable future - what is the group's sense of the likelihood of significantly improved catalysts for direct methanol systems?
- Where can more theoretical approaches to these problems be beneficial? Or will theory simply help explain why the system works once it does work?

NATICK'S FUEL CELL REQUIREMENTS FOR THE SOLDIER SYSTEM

**Brad S. Laprise
US Army Natick RD & E Center**

The Generation II Soldier System Advanced Technology Demonstration (GEN II ATD) is the core element of the 21st Century Land Warrior Integrated Technology Program (21CLW ITP). The focus of the 21CLW 6.3a ITP is to link the individual soldier to the digitized battlefield, establish a command and control network, and provide total situational awareness and near real time automated targeting. The predecessor to 21CLW was the Soldier Integrated Protective Ensemble (SIPE) ATD, the first soldier system oriented research and development effort (FY90-93). SIPE was a modular, integrated fighting system for the dismounted infantryman designed to enhance combat effectiveness while providing balanced multiple threat protection. These technologies provided major improvements in individual/collective performance at night, in an obscured environment and in a chemical environment by enhancing lethality, command and control, survivability and mobility.

The GEN II ATD is directly linked to and fully coordinated with the following 21CLW ITP programs:

FO/FAC	Forward Observer/Forward Air Controller ATD
OICW	Objective Individual Combat Weapon ATD
MPIM	Multi-Purpose Individual Munition TD
ISM	Integrated Sight Modules TD
CIMMD	Close-In Man-Portable Mine Detector ATD
AI ²	Advanced Image Intensifier TD
HRDS	High Resolution Display System TD

The thrust of this effort is the infantry soldier in a temperate environment. The GEN II Soldier System will develop an integrated head to toe individual fighting system. The goals of the GEN II ATD are to show improvements in lethality, survivability, command and control, and mobility in demonstrations in FY96 and FY98. The users include the U.S. Army Infantry School, U.S. Army Signal School, U.S. Marine Corps, and the U.S. Special Operations Command.

The GEN II Soldier System will consist of the following five major subsystems:

IHS (Integrated Headgear Subsystem) - This subsystem will include communications, a helmet mounted display for viewing computer/sensor outputs, an integrated night vision device, ballistic protective shell, and Respiratory Protective Component. The IHS will interface with either/both the OICW or the Modular Weapon System.

ISC/R (Individual Soldier's Computer/Radio) - This subsystem will present information via a color hand held display and/or IHS helmet mounted display, data storage and transfer, mapping capability and video capture/transfer. It will also support voice communications, mine detection, chemical detection, combat identification and a personal status monitor.

WIS (Weapon Interface Subsystem) - This subsystem will consist of interfaces with the OICW and Modular Weapon System although the GEN II Soldier System must be compatible with additional Light Infantry weapon systems used by the soldier.

MCCS (Microclimate Cooling Subsystem) - This subsystem will remove 300 watts of metabolic heat from the soldier for four hours.

PS (Protective Subsystem) - This subsystem will provide multiple threat ballistic protection to selected areas of the torso and appendages in a modular design at a 20% weight reduction. It will also provide multi-spectral signature reduction and advanced load bearing capability.

Of the five subsystems, the Protective Subsystem is the only one which does not require power. While the remaining four do require power, the MCCS, which must remove 300 watts of metabolic heat from the soldier for four hours at a weight of ten pounds, is, by far, the largest consumer. Taking inefficiencies of the system into account and assuming a COP of 2.5, the power requirement of this subsystem is approximately 150 watts (600 watt-hours). Thus, it is evident that the requirements for the MCCS are extremely challenging. A ten pound system weight suggests a four to five pound limit on the weight of the energy source. Presently, we are depending upon batteries to meet this requirement, however, we are turning to fuel cells for technology insertion in the FY98 demonstration.. Since this program will not specifically develop energy sources, we must rely on government technology base efforts, as well as, academia and industry fuel cell experts to help us meet the challenges of the MCCS and the Generation II Soldier System.



Objectives

- **Demo integration of new and existing technologies and tactics to provide total situational awareness and real-time automated targeting for dismounted soldiers**

- **Demo the linkage and integration of the individual soldier to the total force through the digitized C2 network, increasing survivability, lethality and controlled dispersion**

- **Leverage commercial industry to develop a lightweight, rugged, modular and affordable soldier system**



Program Components

- Gen II Soldier System ATD (System Contract)
 - Integrated Headgear Subsystem
 - Individual Soldier Computer/Radio Subsystem
 - Weapons Interface Subsystem
 - Protective Subsystem
 - Microclimate Cooling Subsystem
- Supporting Programs
 - Objective Individual Combat Weapon (OICW) TD
 - Integrated Sight Modules (ISM) TD
 - Advanced Image Intensifier (AI2) TD
 - High Resolution Display System (HRDS) TD
 - Multi-Purpose Individual Munition (MPIM) TD
 - FO/FAC ATD (USMC)
 - Individual Mine Detection TD
 - Combat ID TD
 - Individual Soldier Power TD



Key Milestones - GEN II System Contract

- **Contract Award - 4QFY94**
- **Phase I: Concept Development - 6 Months**
- **Phase II: Critical Component Breadboard Design and Demo - 12 Months**
 - AWE at DBBL to assess/validate compatibility with C2 protocols and architectures
- **Phase III: Subsystem/System Development and Prove-Out (2 systems) - 14 Months**
 - AWE at DBBL to assess/validate overall system operational performance/compatibility and to link "live" GEN II Soldier with constructive and virtual simulations
- **Phase IV: Final System Fabrication & Assembly - 12 Months**
 - Use of constructive/virtual simulations to complete training program, i.e., "how to fight" the system
- **Phase V: Training & Field Demo - 8 Months**
 - AWE at DBBL is the Top Level Demo
 - Platoon-level demo with comms up to Division
 - Live vignettes for training/Jata collection iterated with same vignettes conducted in virtual simulations
 - Culmination in free-play force-on-force live exercise (ala JRTC) (Tentative)

GEN II SOLDIER SYSTEM

Subsystems	Power (W)
1. Integrated Headgear Subsystem (IHS)	10
2. Individual Soldier's Computer/Radio (ISC/R)	25
3. Weapon Interface Subsystem (WIS)	15
4. Protective Subsystem (PS)	
5. Microclimate Cooling Subsystem (MCCS)	150

21st Century Land Warrior

Top Level Demonstration (21CLW TLD)

OBJECTIVE: Demonstrate the enhanced survivability and lethality of the dismounted soldier through:

- Total situational awareness & real time automated targeting
- Linkage to the digitized command & control network
- Enhanced multiple threat protection
- Leverage of the commercial microelectronics & telecommunications industries

SCHEDULE: 1994 - GEN II Soldier system contract award

1995 - Concept Development

1996 - Interim demo to assess critical components and C² viability and compatibility

1997 - System Development/Fabrication

1998 - Top Level Demonstration

APPROACH: The 21CLW TLD is operationally focused on the U.S. Army Infantry, the U.S. Marine Corps (Infantry) and the U.S. Special Operations Forces. The 21CLW TLD builds on the Soldier Integrated Protective Ensemble ATD (SIPE, 1989-1992) and integrates numerous ATDs and TDs of which the Generation II Soldier ATD is the cornerstone.

Weapons Interface Subsystem will link the Objective Individual Combat Weapon and other weapons to the Gen II System.

Individual Soldier's Computer/Radio Subsystem (2-4 lbs) is the link to the digitized C² network and provides real time information through:

- Secure, voice controlled intra and inter squad voice/data communications
- GPS & Inertial navigation, digitized maps and overlays
- Information management (e.g. call for fire)
- Video capture (thermal and daytime)
- Link to sensors: Combat ID

Personnel status monitor
Chemical agent detector
Mine detection

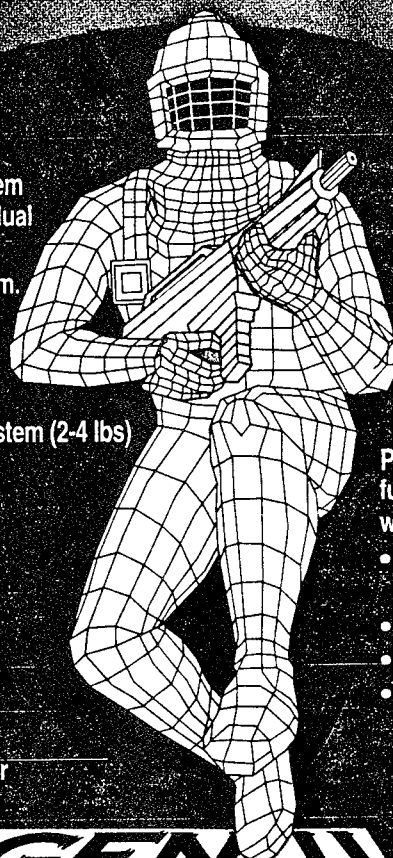
Integrated Headgear Subsystem (5 lbs) views output of the weapon, computer and sensors, and consists of:

- Lightweight ballistic shell
- High resolution helmet mounted display
- Individual soldier imaging system
- Communications interface
- Respiratory protective component

Protective Subsystem will provide the full range of threat protection at 20% reduced weight, in a modular subsystem:

- Multi-threat ballistic protection for the torso and limbs
- Multi-spectral signature reduction
- Enhanced chemical/biological protection
- Advanced load carriage

Microclimate Cooling Subsystem will deliver 300w of cooling for 4 hours in a modular subsystem weighing under 10 lbs.



Integrated Sight Modules TD (CECOM)

Close-In Mine Detection ATD (CECOM)

Ltwt Power Technology (ARL)

Forward Observer/Forward Air Controller ATD (USMC)

Objective Individual Combat Weapon TD (ARDEC)

Modeling & Simulation (DBBL/Natick/ARL TRAC-WSMR)

High Resolution Display System TD (CECOM)

Multi-Purpose Individual Munition TD (MICOM)

The Force of the Future

FOR MORE INFORMATION CONTACT : Carol J. Fitzgerald, 21CLW TLD Manager, SARD-TT, DSN 225-1447, (703) 695-1447 OR Patrick R. Snow, Jr., GEN II ATD Manager, USA Natick RD&E Center, DSN 256-5436, (508) 651-5436

SILENT ENERGY SOURCE FOR
TACTICAL APPLICATIONS
(SIESTA)

Walter G. Taschek
U.S. Army CECOM
Fort Belvoir, VA

The Soldier System includes all items/equipment that are worn, consumed or carried by the soldier in the field for his or her individual use. The development of a power source for the Soldier System is critical if the required capabilities are to be obtained. The inherent characteristics of fuel cell power sources, such as silence and high efficiency, make them prime candidates for a Soldier System power source.

Projected power requirements for the Soldier System vary from approximately 50 watts to 500 watts. This range falls largely in a "Gray Area" for portable power sources (see figure); for low power or short duration missions, batteries are suitable. At combinations of higher power or longer duration missions, high energy density batteries of today are too heavy and costly. Engine generators are not suited to some applications due to signatures.

The U.S. Army Belvoir Research, Development and Engineering Center now CECOM-C2SID identified the need for small "Silent Energy Sources for Tactical Applications" (SIESTA) in the late 1970's. SIESTA was intended to complement power sources being developed under the "Silent Lightweight Electrical Energy Plant" (SLEEP) program by providing power sources rated from 10 to 300 watts. While the SLEEP program was based on methanol fueled phosphoric acid fuel cells in the 1.5-5.0 kW range (4, 5) the SIESTA program was oriented on using metal hydride fuels and PEM fuel cells. Prototypes ranging from three watts to over 30 watts were successfully demonstrated using calcium hydride fuel. When the SLEEP program was ended due to logistical impacts associated with the use of Methanol fuel, work on all fuel cell programs at Belvoir ended.

The state-of-the-art has evolved significantly since the 1970's. In particular the Proton Exchange Membrane (PEM) has progressed to a point where major application efforts are underway. The inherent advantages of the PEM Fuel Cell make it a prime candidate for individual power source applications. These include: low operation temperature; high performance levels; and an excellent electrolyte material (6). The low operating temperature of less than 100 deg C provides for low thermal signature and rapid startup. It also decreases the thermal stress, which is important since the military units will be subjected to hundreds of start-stop cycles. The Army Research Office has sponsored PEM fuel cell efforts at Analytic Power Corporation aimed at producing lightweight stacks in the power range of the Soldier System requirements. The Advanced Research Projects Agency (ARPA) also has PEM fuel cell programs, aimed at the direct utilization of liquid hydrocarbon fuels.

The required capabilities for the individual soldier depend on the type of soldier and the mission. The Soldier System will require innovative power sources to meet requirements. The soldier system of the future will include a wide variety of power consuming equipment. In general, the electronic devices are low power consumers. Their required power is currently estimated at 50 watts. Batteries are adequate power sources for most missions at this power level. The power for the microclimate control equipment is 150 watts based on a COP of 2. The current requirement for the dismounted combat soldier is a 72 hour mission without resupply, including 12 hours of operation in a Chemical/Biological contaminated environment. This leads to a power source reasonably sized at 150 watts that will provide 1800 watt-hours. Total allowed weight of the microclimate system and power source is 11 pounds. A program has been initiated to demonstrate a system with a six hour mission by 1996 (900 WHe) and an eight hour mission by 1998 (1200 WHe) as milestones on the path to the desired mission of twelve hour operation over a 72 hour period (1800 WHe).

There also are desired capabilities, not as well defined at lower and higher power. These include exoskeletal devices, weapons, and power tools. Refuelable power sources are required for high energy missions. Just as there is no one-size engine generator set in the field that will meet all mission requirements, several power sources for the individual soldier also will be required to handle the variety of soldier missions.

One of the key issues associated with the PEM fuel cell work is the source of hydrogen. Hydrogen storage/production is being investigated by almost all of the organizations involved in fuel cell research and development. The storage technologies include pressurized gas, liquid hydrogen, and hydrides (disposable and rechargeable). The challenge is to supply the hydrogen in a safe, lightweight package. The source of oxygen can also be important for military applications. Most systems utilize the oxygen from ambient air, but in an environment containing salt fog or chemical agents some treatment may be necessary to protect the fuel cell.

Improvements in the cell stack technology are expected to continue and the hydrogen source, for the near term is expected to be the pacing item. The ARPA work on hydrocarbon fuels could offer even better systems in the mid and long term.

The Soldier System application will require a power source that is silent, low in signature, able to operate in any attitude, safe, and cost effective. It must not interfere in any way with the soldier's ability to perform the mission. Ease of operation and simplicity of design are crucial to the success of a fuel cell system for this application.

**SILENT ENERGY SOURCES FOR
TACTICAL APPLICATIONS
(SIESTA)**

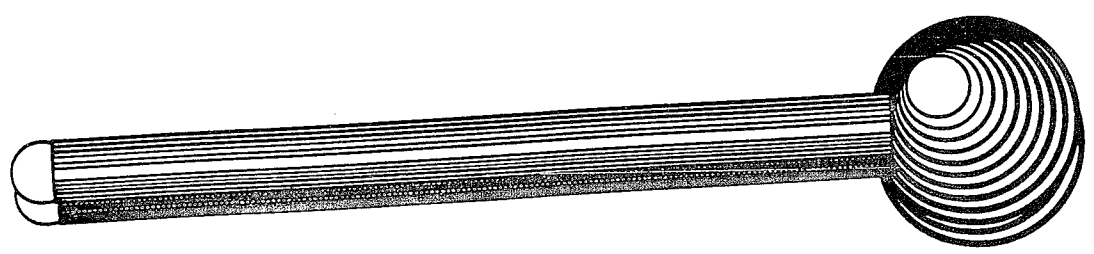
PROSPECTOR VII

**WALTER G. TASCHEK
CECOM-C2SID-SOUTH
FORT BELVOIR, VA**

1 NOV 94

Fuel Cell Technology

Operating Temperature (C)



1000

900

800

700

600

500

400

300

200

100

0

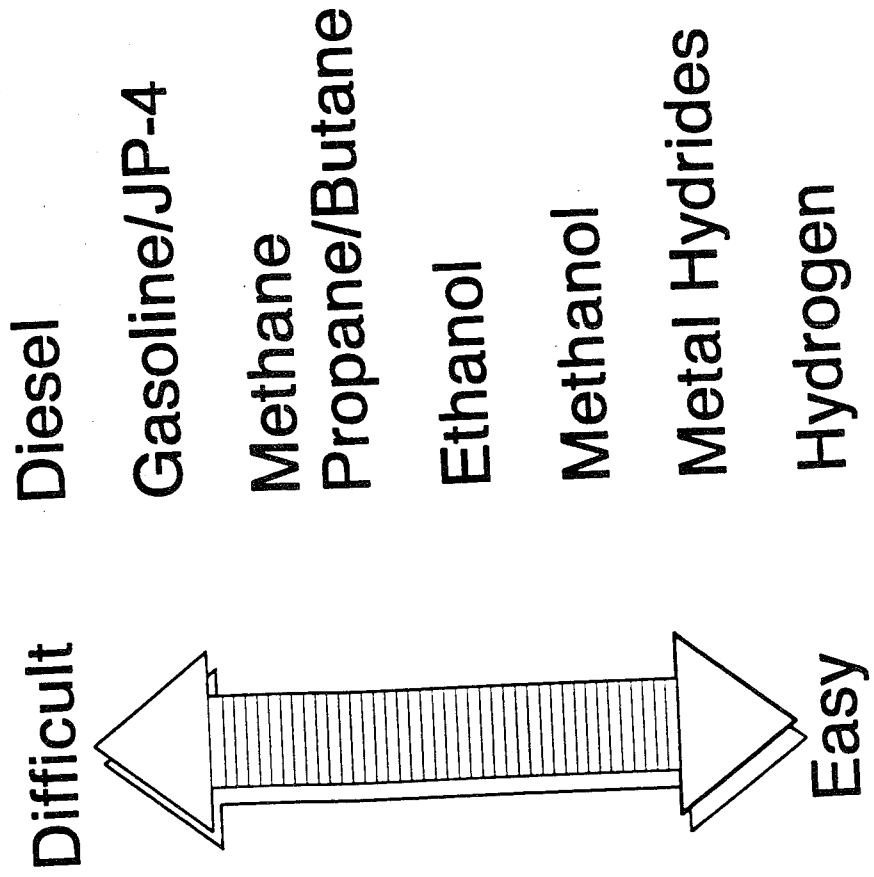
Solid Oxide

Molten Carbonate

Phosphoric Acid

PEM

FUEL PROCESSING



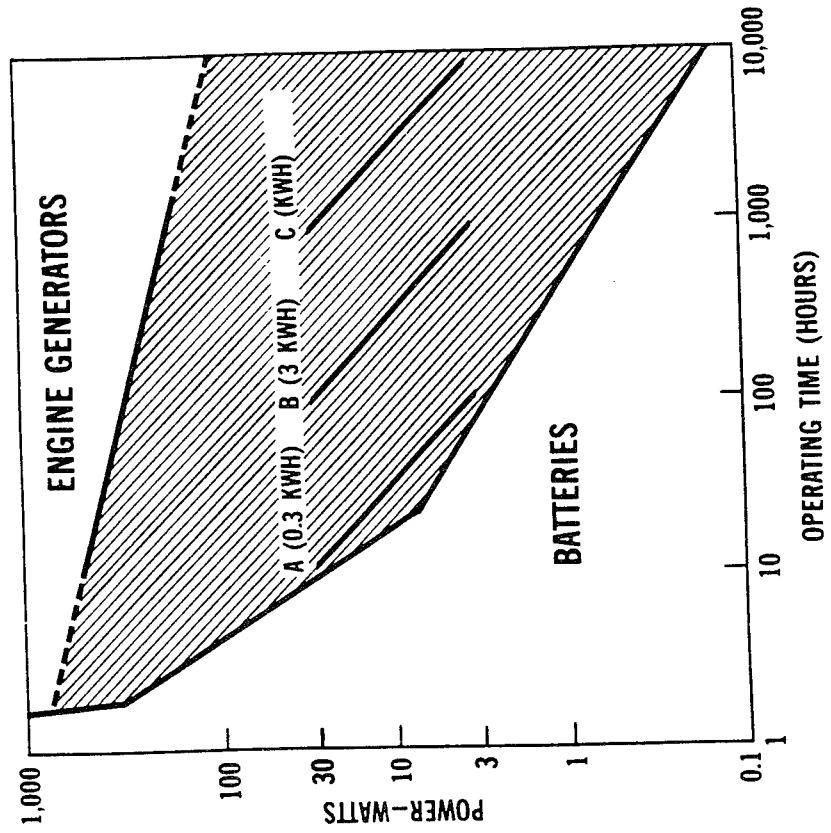
PEM AND SMALL FUEL CELL R&D AT BELVOIR RD&E CENTER

- 1966-69 300 Watt Hydrazine Fuel Cell
- 1972 Milliwatt Hydride Fueled Phosphoric Acid (HPA) Demos
- 1973-74 REMBASS XRAY HPA Power Source (2-12 Watt)
- 1975 GE 12 Watt SPE Stack Evaluation
- 1975 Miniature Hydrogen Generator
- 1976 Contract to Engelhard for 30 each 3 Watt SPE (PEM)
Stack DAAK 53-76-C-0130

PEM AND SMALL FUEL CELL R&D AT BELVOIR RD&E CENTER (continued)

- 1976-77 Landing Light Demos (approximately 12) 3 Watt PEM Stack
& Miniature Hydrogen Generator
- 1977-78 Contract for 30 Watt, 28 VDC, Hydride PEM Fuel Cell
(DAAK70-77-C-0222). Included an additional two
30 Watt PEM Fuel Cell Stacks for DREO.
- 1977-78 Evaluation and Optimization of SPE Fuel Cells Using
Reformate Fuels. Contract DAAK70-77-C-0218, GE
- 1978-79 Component Development & Performance Mapping of PEM
Fuel Cells at Environmental Extremes. Continuation of
Contract DAAK70-77-C-0222 with Engelhard.
- 1979-82 Silent Energy Sources for Tactical Applications (SIESTA)

BRIDGING THE GRAY AREA



MISSION WEIGHT - POUNDS

	30 WATT F.C.	BB 501
A	15	32
B	30 (23)	320
C	165 (90)	3200

() NOT INCLUDING WATER

SCIENCE AND TECHNOLOGY OBJECTIVE

IV.H.3

LIGHTWEIGHT FUEL CELL TECHNOLOGIES

FOR

INDIVIDUAL SOLDIER POWER NEEDS

BY FY96

- * INTEGRATED FUEL CELL POWERED MCC WEIGHING 7Kg - 300 WATTS METABOLIC COOLING FOR 6 HOURS
- * BATTERY CHARGER PROVIDING 1200 WATT-HOURS OF CHARGING PER Kg OF FUEL
- * EVALUATION OF HIGH POWER DENSITY PRESSURIZED PROTON EXCHANGE MEMBRANE FUEL CELL STACKS.

BY FY98

- * INTEGRATED FUEL CELL POWERED MCC WEIGHING 5Kg - 300 WATTS METABOLIC COOLING FOR 8 HOURS

SOLDIER SYSTEM POWER CONSUMERS

72 HOUR MISSION

<u>SUBSYSTEM</u>	<u>USAGE (HOURS)/POWER</u>	<u>ENERGY</u>
INTEGRATED HEAD GEAR	18 / 8 WATTS	144 WATT-HOURS
COMPUTER/RADIO	36 / 10 WATTS	360 WATT-HOURS
WEAPON INTERFACE	24 / 10 WATTS	240 WATT-HOURS
MICROCLIMATE COOLING	12 / 150 WATTS	1800 WATT-HOURS

PROJECTED POWER AND ENERGY REQUIREMENTS

Duration	Power - Watts	Energy - W-Hrs
4 Hours	300	1,200
6 Hours	30	180
8 Hours	100	800
24 Hours	100	2,400
24 Hours	500	2,000
48 Hours	250	12,000
72 Hours	300	5,000
7 Days	100	700
30 Days	50	3,000

TYPES OF SOLDIERS

- **Transportation**
- **Military Police**
- **Engineer**
- **Ordnance**
- **Intelligence**
- **Chemical**
- **Signal**
- **Quartermaster**

POWER CONSUMING EQUIPMENT

- Night Vision Devices
- Thermal Imaging Devices
- Visual Helmet Display
- Communications Equipment
- Individual Computer
- Enhanced Hearing
- Global Positioning System
- Weapons Ranging System
- Microclimate Control System
- CB Monitor

SOLDIER FUEL CELL POWER POTENTIAL USES

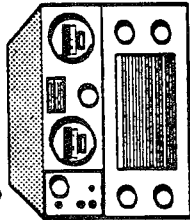
- MICROCLIMATE CONDITIONING
- BATTERY RECHARGE
- PORTABLE POWER SOURCE
- POWER FOR ENHANCED MOBILITY

HYDROGEN SOURCES

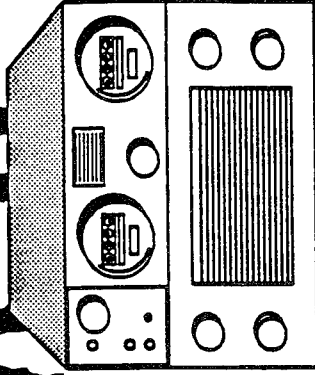
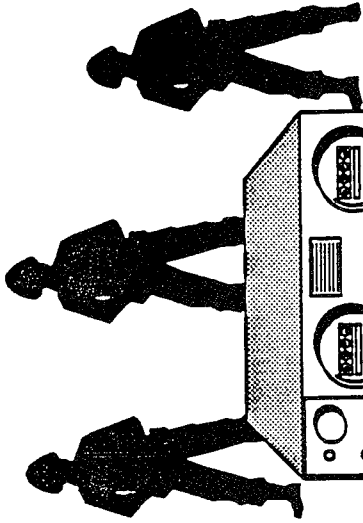
- Rechargeable Hydrides
- Non-Rechargeable Hydrides
- High Pressure Gas
- Liquid Hydrogen
- Hydrocarbon Fuels
- Others

LIGHTWEIGHT GENERATOR

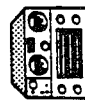
Current Generator Sets Are Heavy



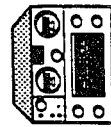
500 Watt
85 Lbs



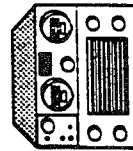
3 KW
285 Lbs



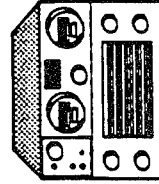
500 W
15 Lbs



1.5 KW
30 Lbs



3 KW
40 Lbs



5 KW
73 Lbs



Our Weight Goals

FUEL CELL POWER SOURCES

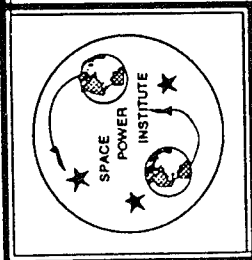
AGREEMENTS/LEVERAGING

AGREEMENTS

CECOM/NATICK PLAN FOR THE DISMOUNTED SOLDIER SYSTEM
CECOM/ARL TECHNOLOGY PROGRAM ANNEX - FUEL CELL RESEARCH
CECOM/ARO MOU - MAN PORTABLE FUEL CELLS
CECOM/ARPA MOU ON FUEL CELL PROGRAM

LEVERAGING

NSA - SMALL FUEL CELL POWER SOURCES
ARPA - DIRECT FUEL CELLS/SOFC
ARL - FUEL CELL COMPONENTS
ARO - FUEL CELL STACK TRANSITION
NATIONAL FUEL CELL COORDINATING GROUP
ABCA - DATA INTERCHANGE
DOE - PEM TECHNOLOGY FOR TRANSPORTATION
IAPG - FUEL CELL TECHNOLOGY



PROSECUTOR BRIEF

FUEL CELLS -- AN UPDATE ON DOD, ARPA AND COMMERCIAL RESEARCH

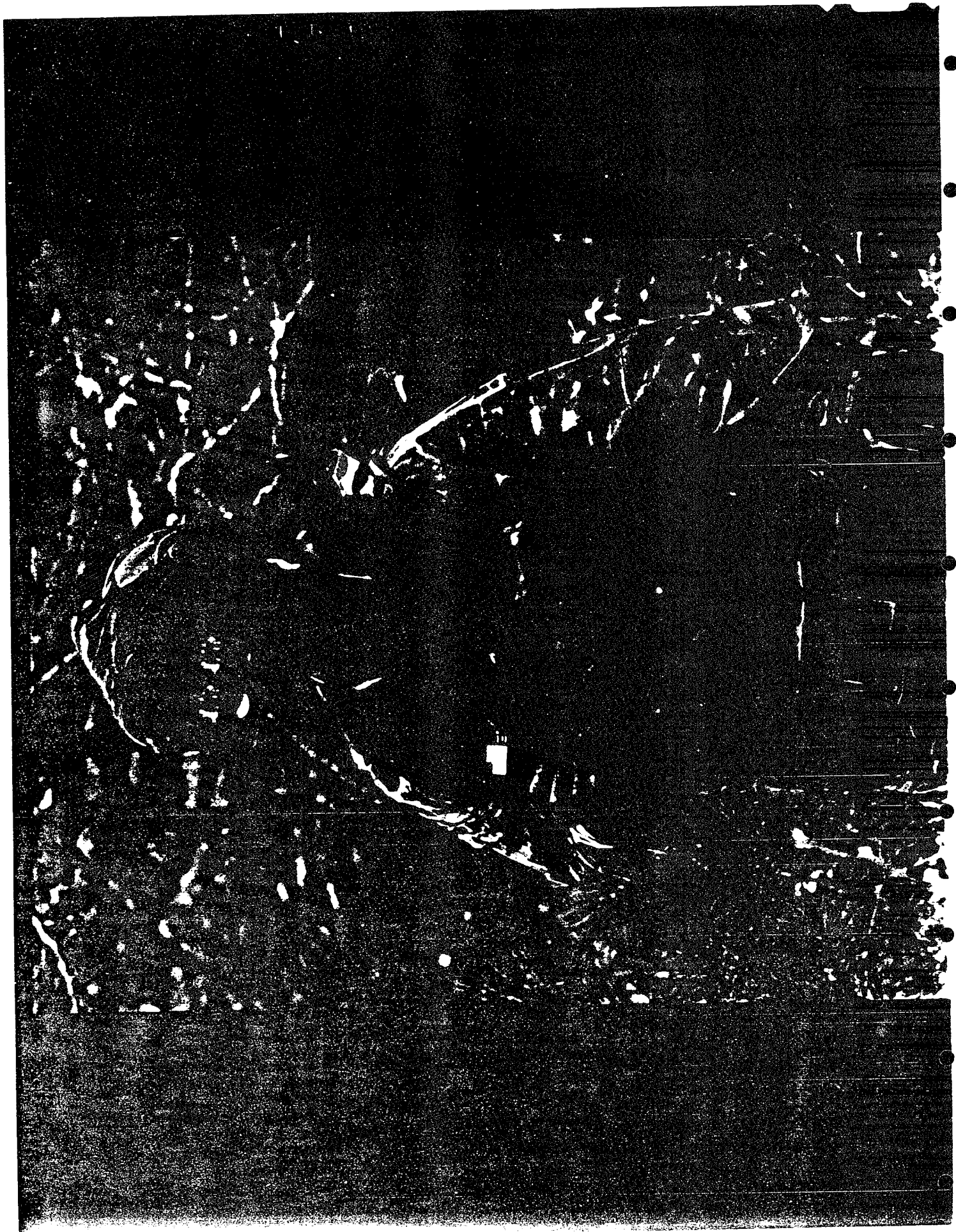
WASHINGTON DUKE INN
OCT 31-NOV 3, 1994

Sponsored by
The Army Research Office

"COMPETING TECHNOLOGIES FOR POWER GENERATION"

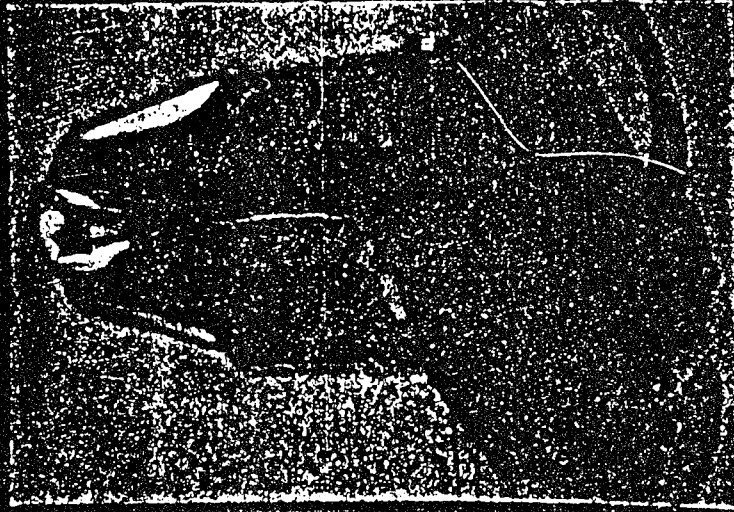
M. Frank Rose
Auburn University

19 - 0 ain't bad !!War Eagle!!



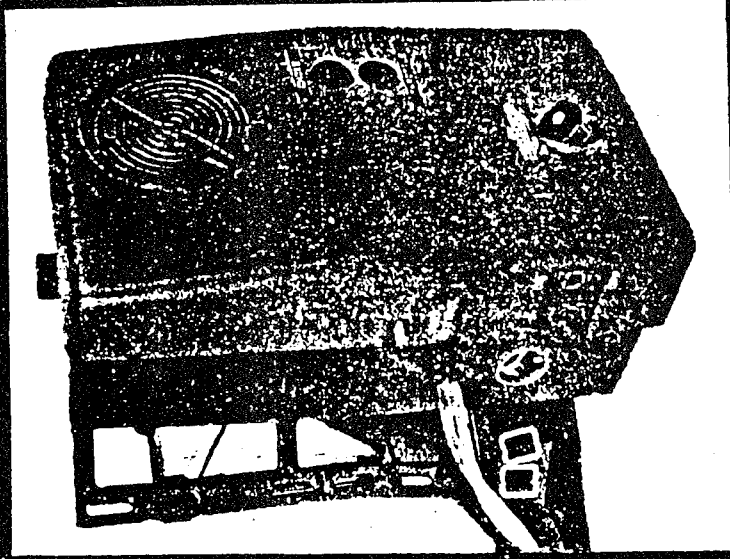
INDIVIDUAL MICROCLIMATE COOLING

1986



Weight: 30.0 pounds
Volume: 0.90 cubic feet
Fuel: GASOLINE
Mission: 4-6 HRS

1992



Weight: 16.7 pounds
Volume: 0.55 cubic feet
Fuel: JP-8
Mission: 4-6 HRS

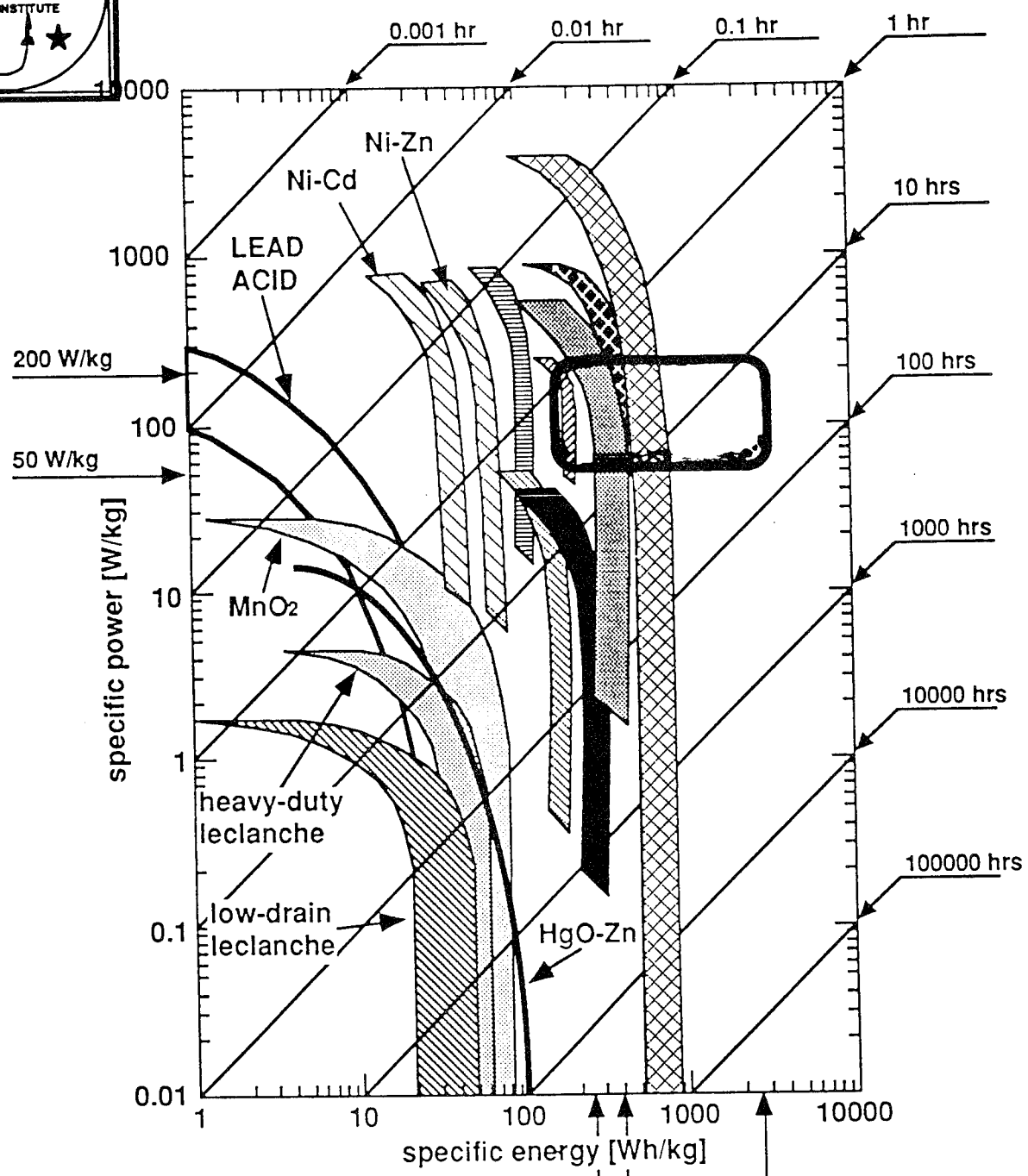
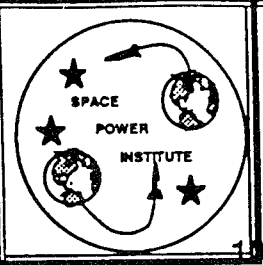
2004



Weight: <10 pounds
Volume: 0.3 cubic feet
Fuel: ?
Mission: 72 HRS

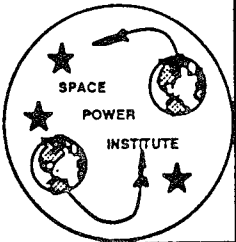
Assess the state-of-the-art of small engines/ power systems

Numerous small engines and advanced concepts were presented at the Workshop. In general, the current state-of-the-art is a specific power on the order of 0.7 hp/in^2 (80.94 W/cm^2) and a specific fuel consumption on the order of 1.2 lb/hp-hr ($7.31 \times 10^{-4} \text{ kg/W-hr}$). It is anticipated that, given sufficient funding, the near term state-of-the-art is on the order of 1.3 hp/in^2 (150 W/cm^2) and a specific fuel consumption of 0.6 lb/hp-hr ($3.66 \times 10^{-4} \text{ kg/W-hr}$). At least one engine which was reported at the Workshop achieved a "best case" fuel consumption of 0.7 lb/hp-hr . These values are consistent with the estimates necessary to power the most demanding of the Soldier System missions. It is impossible to estimate the impact of having to conform to requirements for signature reduction, all orientation operation, cold starting, etc., as discussed in the Workshop proceedings. It was a Workshop consensus that progress is limited by the availability of funds rather than by good ideas directed toward improving small engine technology.



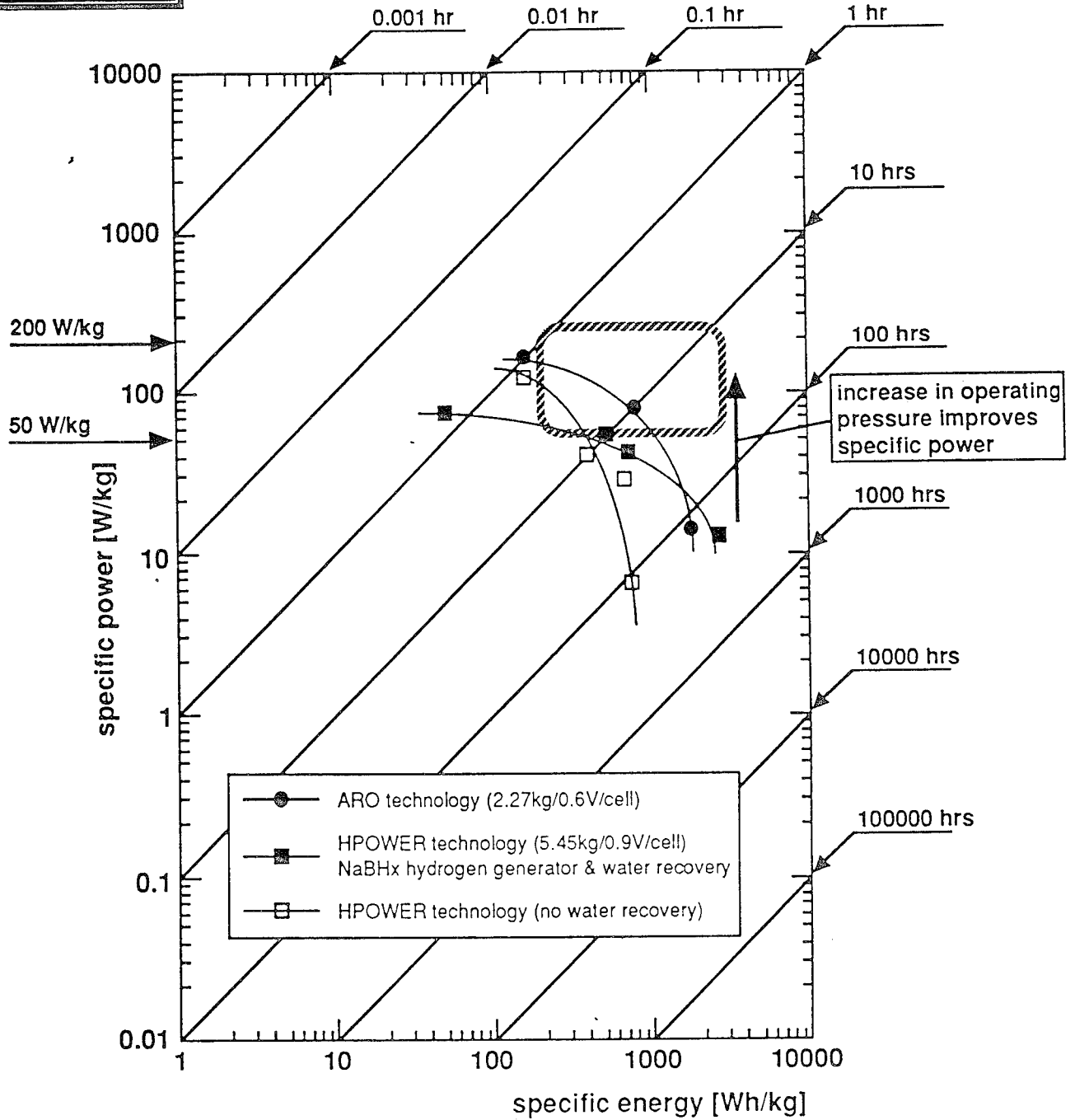
- Soldier System Requirement
- Li/inorganic
- Li/organic
- Li-Cl₂
- Li-S
- Ag-Zn
- Zn-air
- NaS


SPACE POWER INSTITUTE

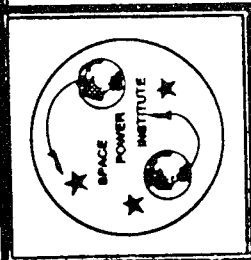


Ragone Plot for Fuel Cell Technologies

(note: mass must always be specified)



 Soldier System Requirement

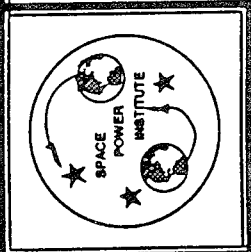


AMTEC

ALKALI METAL THERMOELECTRIC CONVERTER

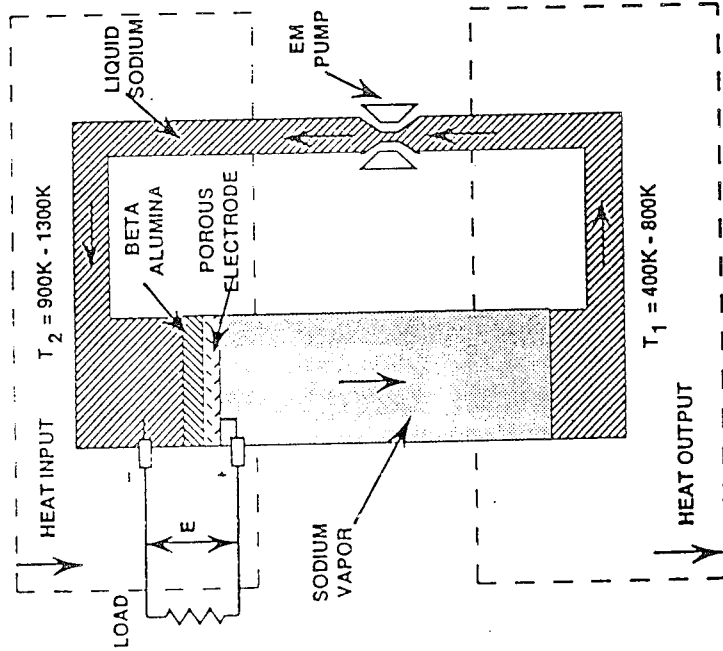
- Army Investigated the Concept Many Years Ago
- Whats New!!!! NASA DoE, Industry and the Japanese have spent Millions of Dollars on R&D
- Efficiencies of 20% have been Demonstrated. Efficiencies Greater than 30 % Feasible. Power Densities as High as 400 W/kg Projected.
- 2000 Hours Continuous Operation at >13% Efficiency.
- "Low" Temperature Operation Possible. Less than 1000 Degrees Centigrade.
- Industrial Interest Insures Availability of Converter Components.
- Technology Ready for Demonstrator in the 10-100 W Range.

AMTEC IS ALMOST READY FOR APPLICATION !!



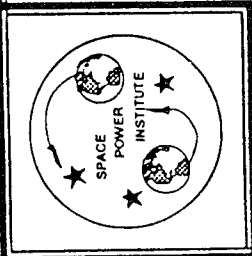
AMTEC

CYCLE SCHEMATIC

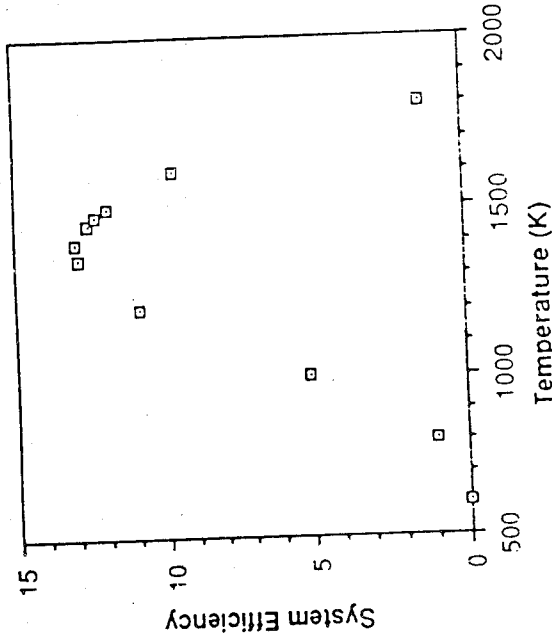
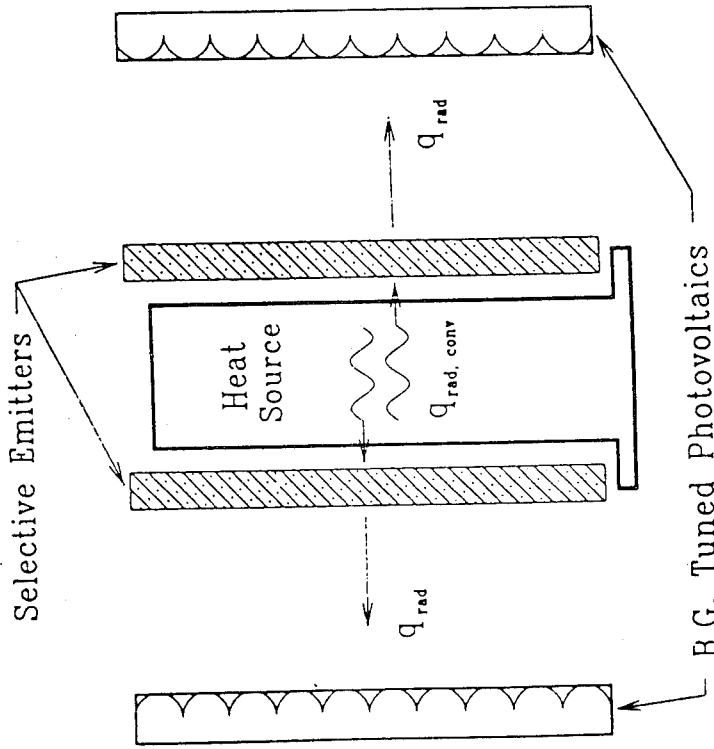


CHARACTERISTICS

- Capable of high efficiency (20-40 percent)
- Heat input at 900-1300 K
- Efficiency independent of size
- Converter power density > 0.5 kW/kg or 0.5 kW/liter
- Modular
- No moving parts (static system)
- Long life potential
- Uses commercially available materials

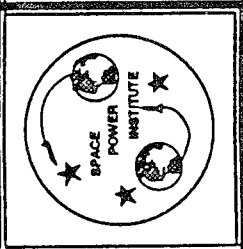


TPV THERMOPHOTOVOLTAICS

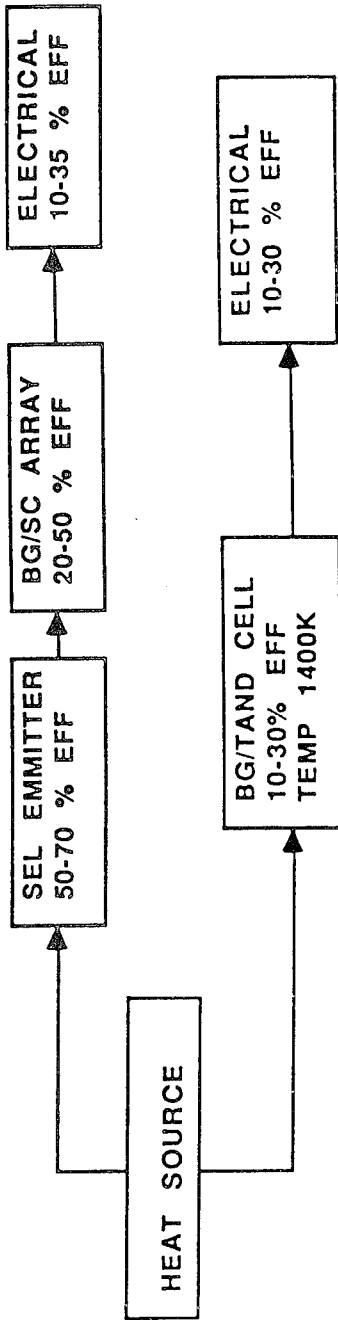


Modeled RTPV System Efficiency vs. Heat Source Temperature. Peak Efficiency Occurs at T=1400K. Note: data assumes cell operating temperature of 330K.

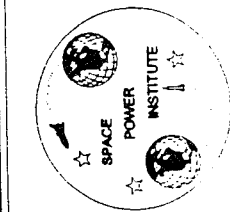
Radioisotope Thermophotovoltaic Power System Utilizing the GaSb IR Photovoltaic Cell
 Mark D. Morgan, William E. Horne, and Phillip R. Brothers, Boeing Defense and Space Group,
 Seattle, WA



TPV THERMOPHOTOVOLTAICS



NOTE!!! DEMONSTRATORS FOR BOTH VERSIONS CAN BE BUILT WITHIN 2 YEARS



Why Are Burners An Issue?



TPV System Efficiency:

$$\eta = \eta_{TS} \eta_E \eta_{PV}$$

Expected Results: $\eta_{TS} \approx 0.9$ Well designed and insulated thermal source

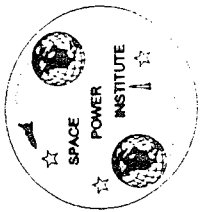
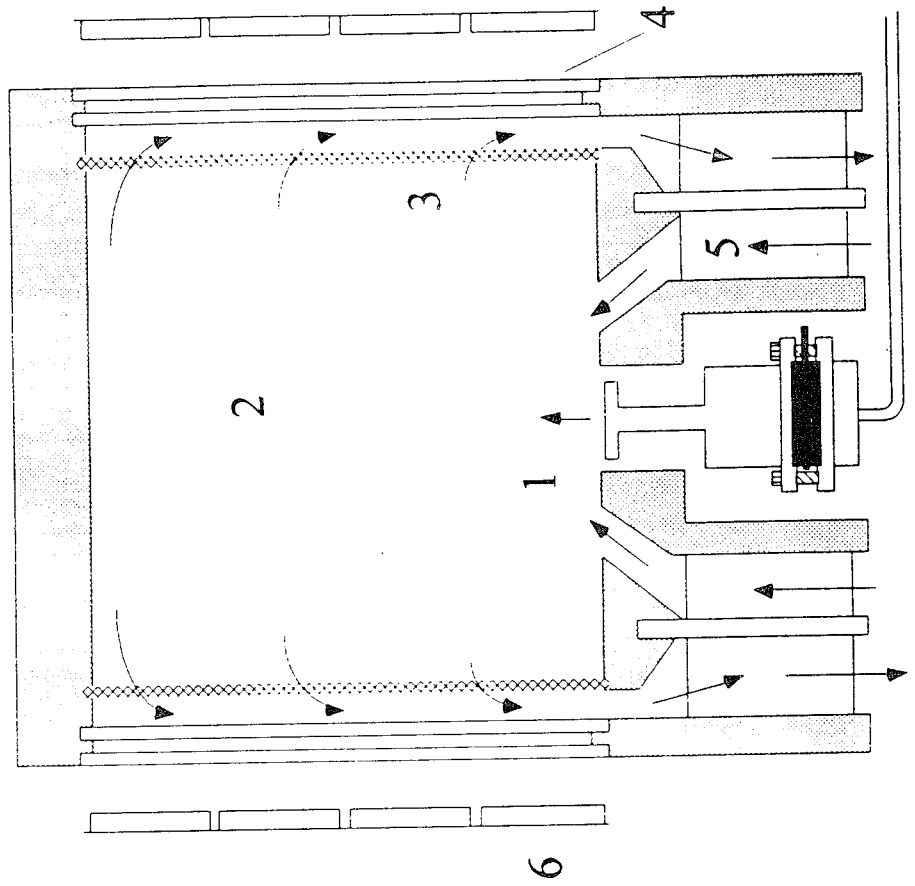
$\eta_E \approx 0.7$ Ho has max $\eta_E \approx 0.7$;

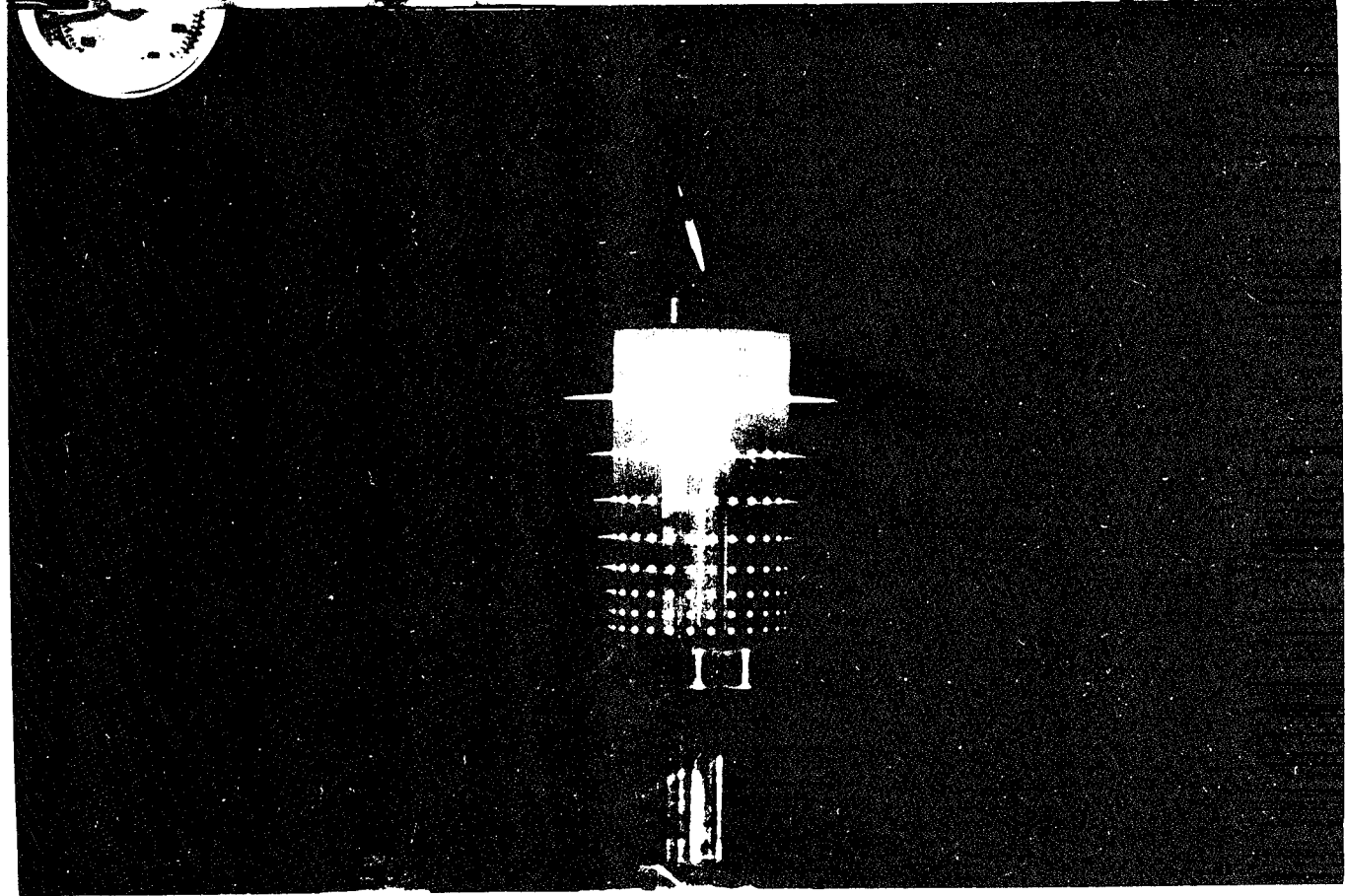
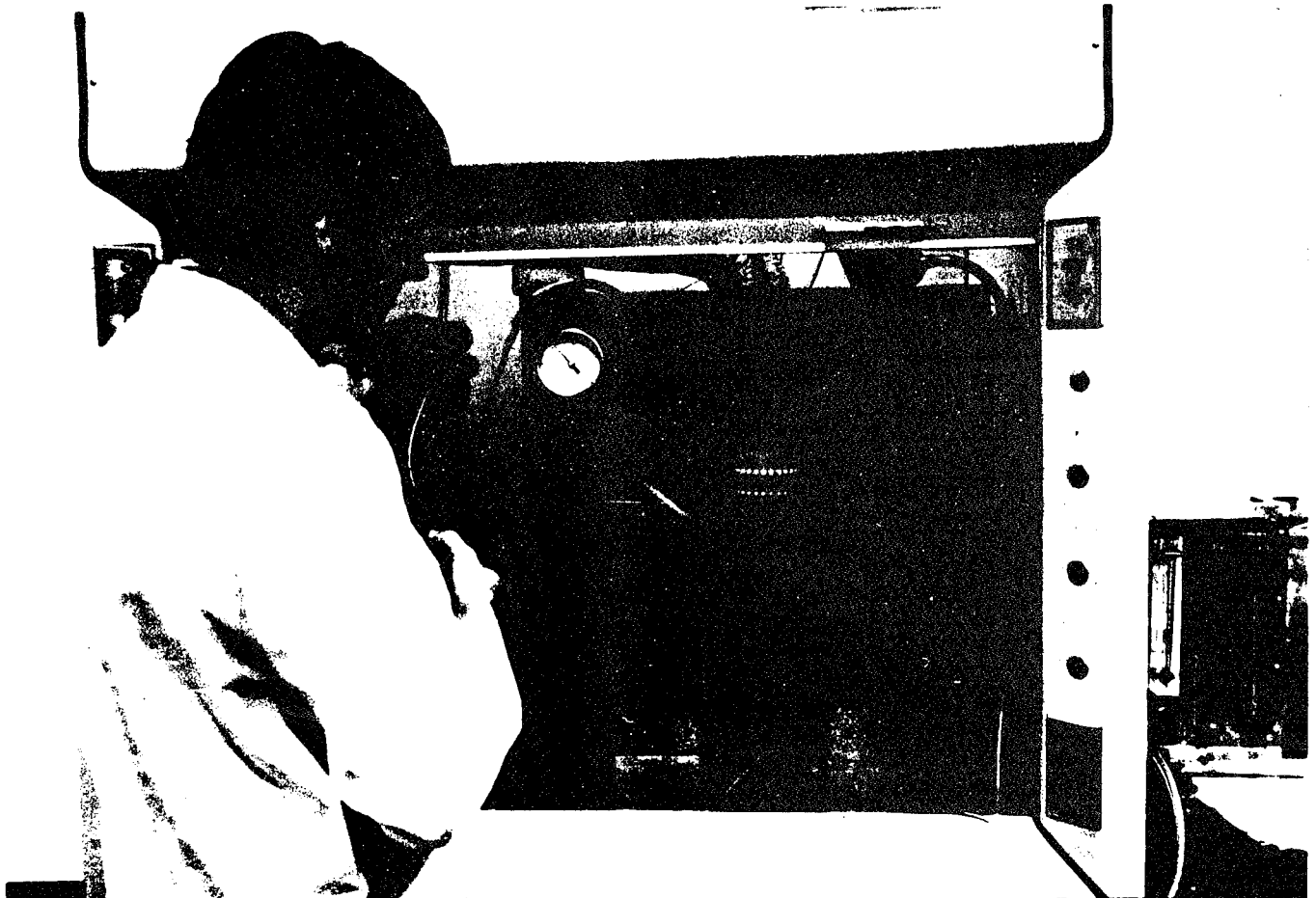
$\eta_{PV} \approx 0.5$ GaAs have shown $\eta_P > 0.5$ measured as Bandgap $E_v = 1$.

Therefore; $\eta \approx (0.9)(0.7)(0.5) \approx 0.3$



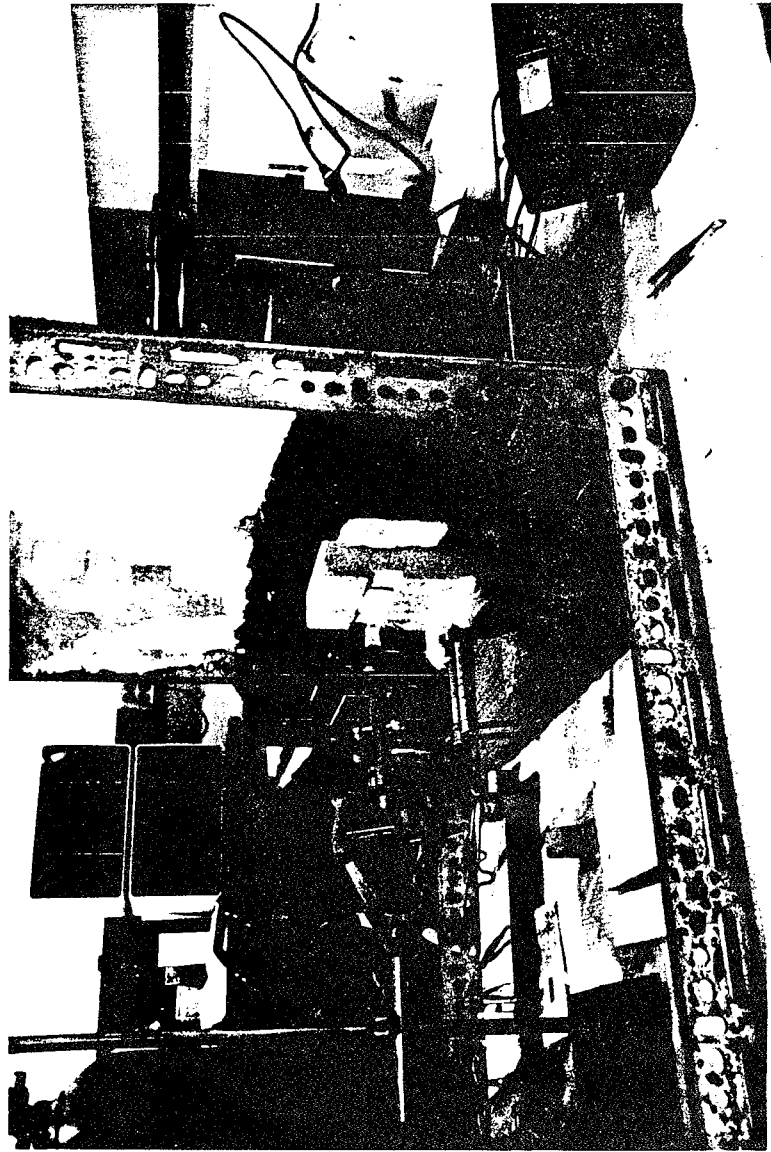
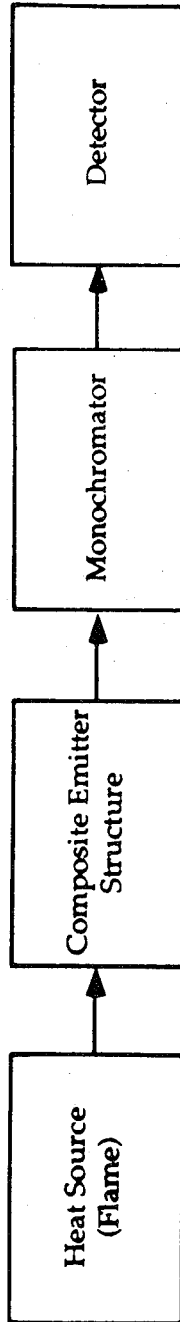
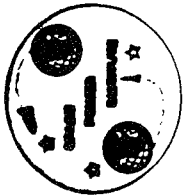
Prototype TPV System

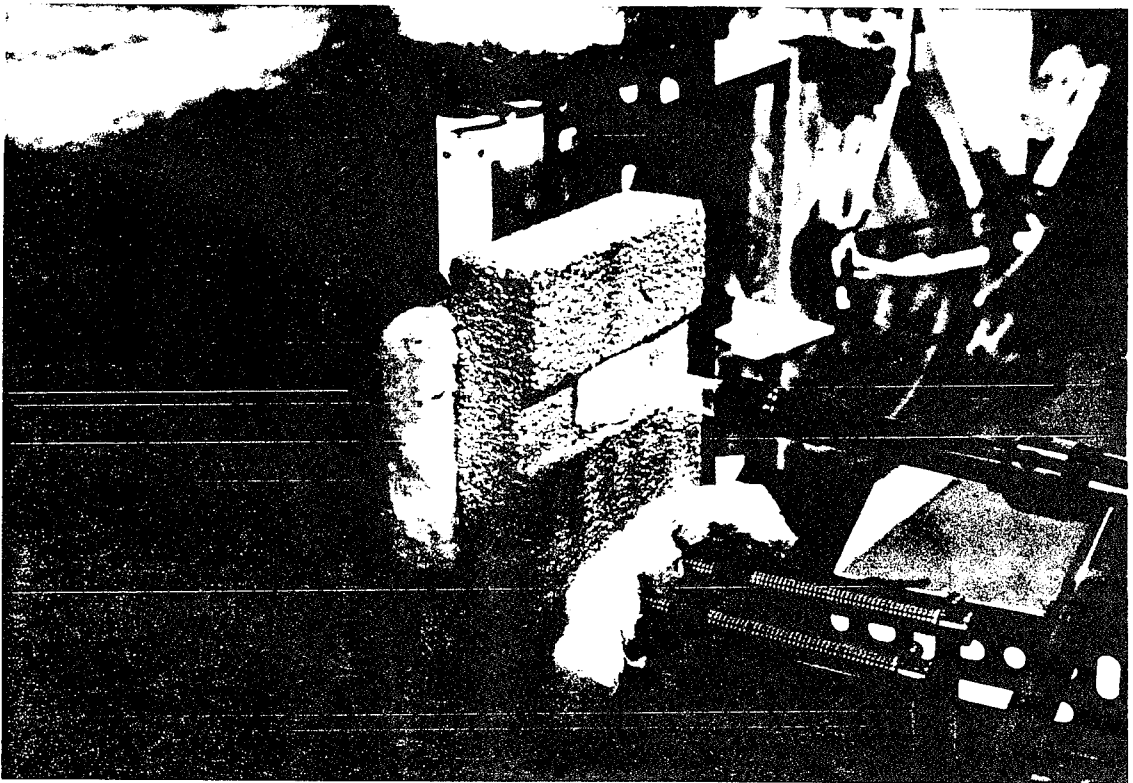
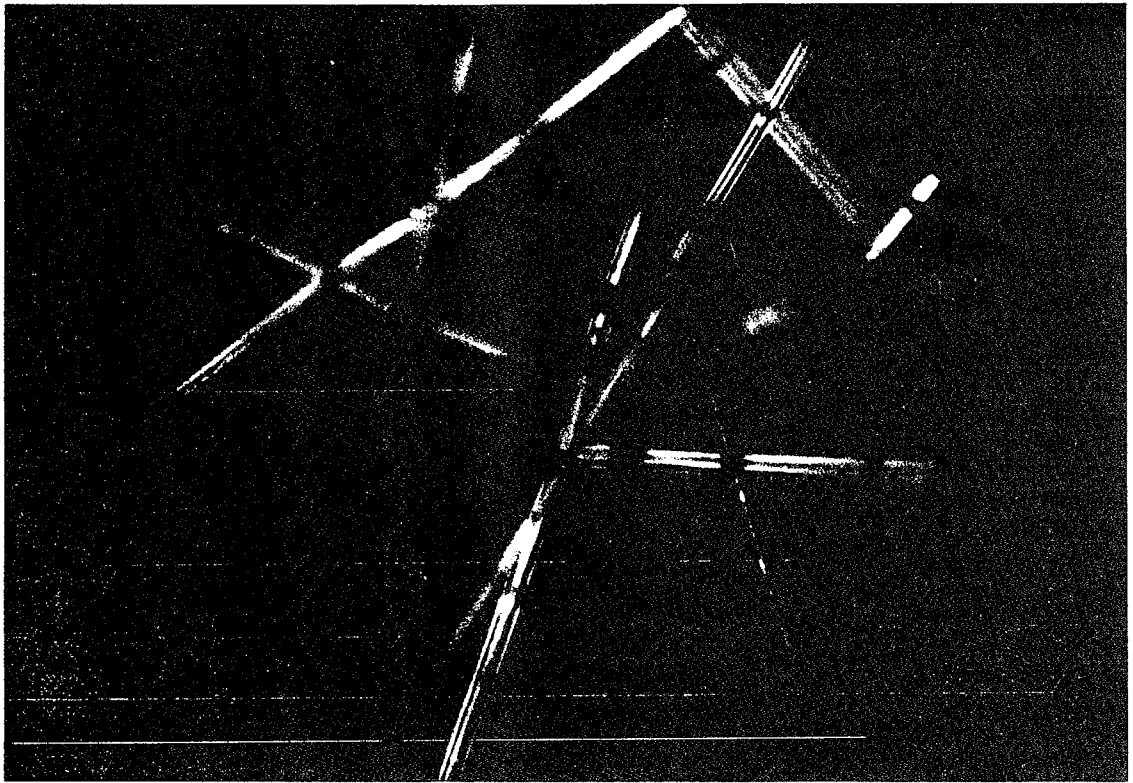


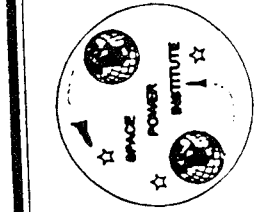




Experimental Setup #1





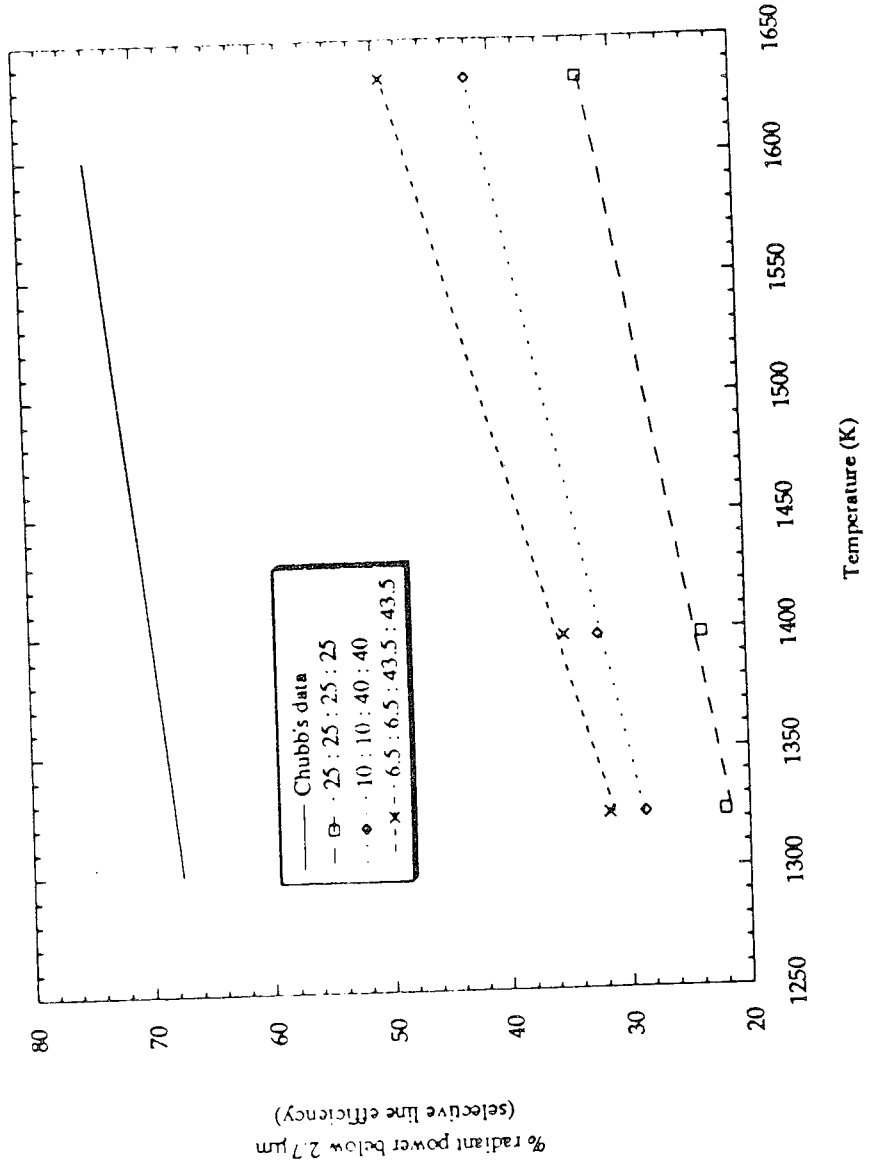
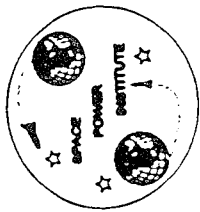


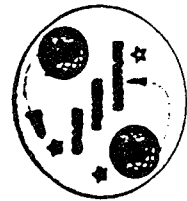
Conclusions

- Composite emitters fabricated which contain silicon carbide or rare earth oxides within strong structural fiber matrix
- Selective line emitters show as much as 50% of radiant power in selective line for erbia and holmia and as much as 33% for ytterbia
- Selective line composite emitters should improve overall TPV system efficiency by lowering the requirements for the heat source



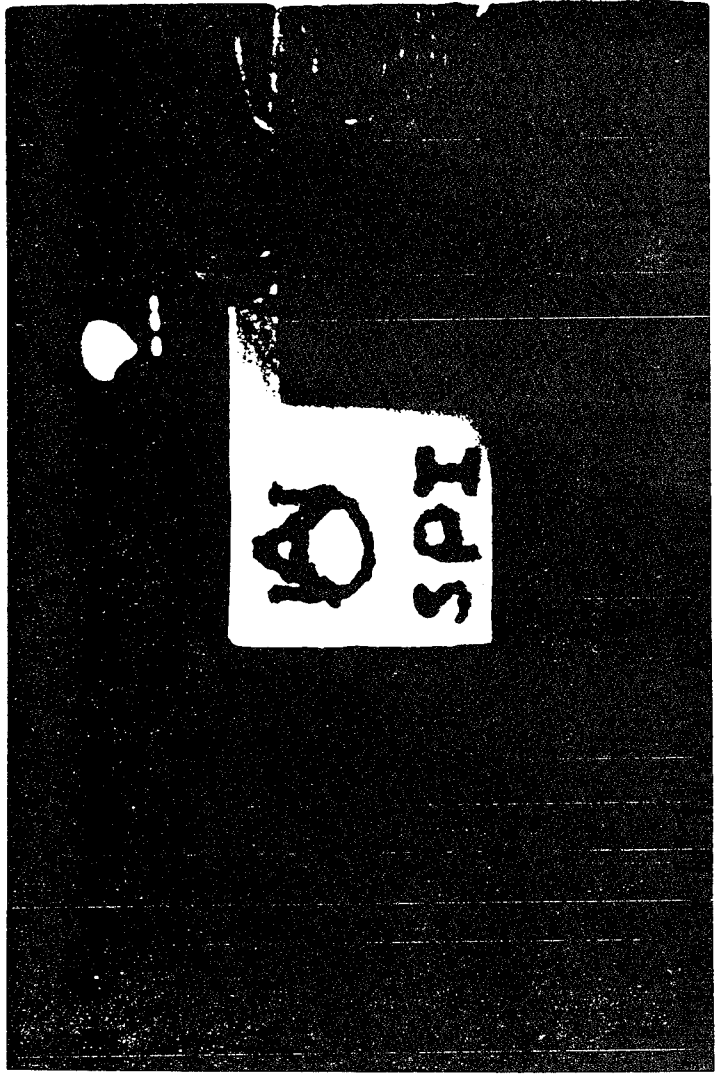
Comparison of Holmia Composite Emitters

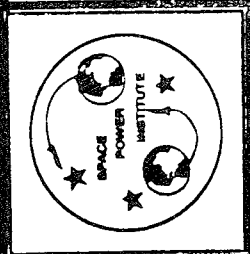




Acknowledgements

This research was sponsored by the Army Research Office under research grant DAALO39260205. The authors would like to thank them for their funding and support.





MOBILE BATTLEFIELD POWER

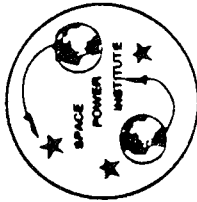
ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO ICE

ASSUMPTIONS

- Soldier must carry one gallon of water (3.79 kg) per day. Total for a three day mission is 11.37 kg
- $T_i = 77K$ $T_f = 300K$ $C_p = 1.84 \text{ J/gmK}$ (77K-277K)
 $C_p = 4.18 \text{ J/gmK}$ (277K-300K) $H_f = 335 \text{ J/gm}$
- From NATICK TR-84/048L, pump, battery, insulation for "ice cooler" is 2.85 kg



MOBILE BATTLEFIELD POWER

ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO ICE

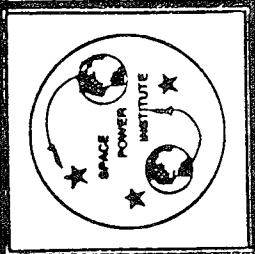
$$E1/M = Cp(T_f - T_i) = 1.84(277 - 77) = 368 \text{ J/gm}$$

$$E2/M = H_f = 335 \text{ J/gm}$$

$$E3/M = Cp(T_f - T_i) = 4.18(300 - 277) = 96.14 \text{ J/gm}$$

$$E_t = 799 \text{ J/gm} = 799 \text{ kJ/kgm} = 222 \text{ watt-hrs/kg}$$

If the soldier needs 300 W for 12 hours, that is 3600 watt-hrs. The cryo ice needed would be 16.2 kg. If the ancillaries weigh 2.85 kg and with a 10% penalty for losses, the pack would weigh 20.65 kg. Subtracting off the 11.37 kg of water the soldier had to carry for drinking, leaves a system net of 9.28 kg - which is competitive



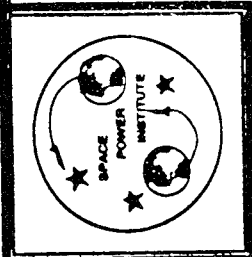
MOBILE BATTLEFIELD POWER

ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO ICE

- Water may be doped with medicine, glucose, or other ingredients selectively
- Cryopack will be segmented and it is possible to produce a vest not requiring a "distinct heat exchanger"
- In order to use the water for drinking, slight modifications to the vest as currently configured will be necessary
- Slightly heavier than currently contemplated for the soldier system
- Minimum R&D necessary in order to reduce to practice



MOBILE BATTLEFIELD POWER

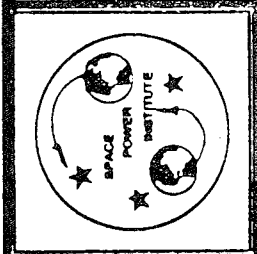
ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO ICE

- By going to 77K for the initial temperature, the cooling capacity is almost tripled when compared to a unit based upon the heat of fusion
- A cooling unit based upon this idea has no thermal, chemical or acoustic signature
- The unit produces drinkable water which is consumed as part of the soldiers consumable allotment
- The soldiers load decreases with time, reducing fatigue when nearing the end of the mission
- There are no environmental or explosives hazards with this approach
- Resupply/recharge is simply replacing cryobricks of water

SPACE POWER INSTITUTE



MOBILE BATTLEFIELD POWER

ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO AMMONIA

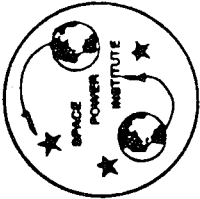
ASSUMPTIONS

- $T_i = 77K$ $T_f = 300K$ $T_{melt} = 196K$ $T_{boil} = 240K$
 $C_p = 2.10 \text{ J/gmK}$ $(85K - 170K)$ $C_p = 2.2 \text{ J/gmK}$ $(240K - 300K)$
 $H_f = 451.86 \text{ J/gm}$ $H_v = 1367 \text{ J/gm}$

- Assume 3 kg for auxiliaries for the heat exchanger, etc.
- Summing as for cryoice, the total energy per gram to raise the temperature of one gram of cryo ammonia from 77K to 300K is:

Eg = 2242 J/gm - roughly a factor of 10 better than water

To meet the soldier system requirement requires 5.7 kg of cryo ammonia at 10% loss, it would require 6.27 kg. Total system mass of approximately 10 kg would appear possible.



MOBILE BATTLEFIELD POWER

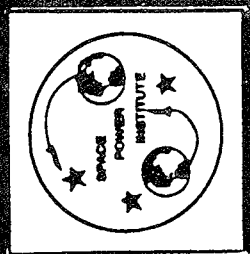
ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO AMMONIA

DISADVANTAGES

- Requires the introduction of ammonia into the battlefield as a fuel
- Ammonia is highly toxic
- Produces an odor which is a strong signature for hostile detection
- Requires high strength confinement after entering vapor phase
- Fuel cell technology in an uncertain state - not clear if an ammonia-air fuel cell would work



MOBILE BATTLEFIELD POWER

ANNUAL REVIEW

CRYOCOOLING FOR THE SOLDIER SYSTEM

CRYO AMMONIA

AMMONIA IS TOXIC

- OPTIONS

- A fuel cell based upon the reaction below is possible



- Simply burn the ammonia producing the same reaction above but release the nitrogen into the air and try to claim the water for drinking

Dubois

**The ARPA/DSO Program In Advanced Fuel
Cell Technology**

Dr. Lawrence H. Dubois

**Defense Sciences Office
Advanced Research Projects Agency**

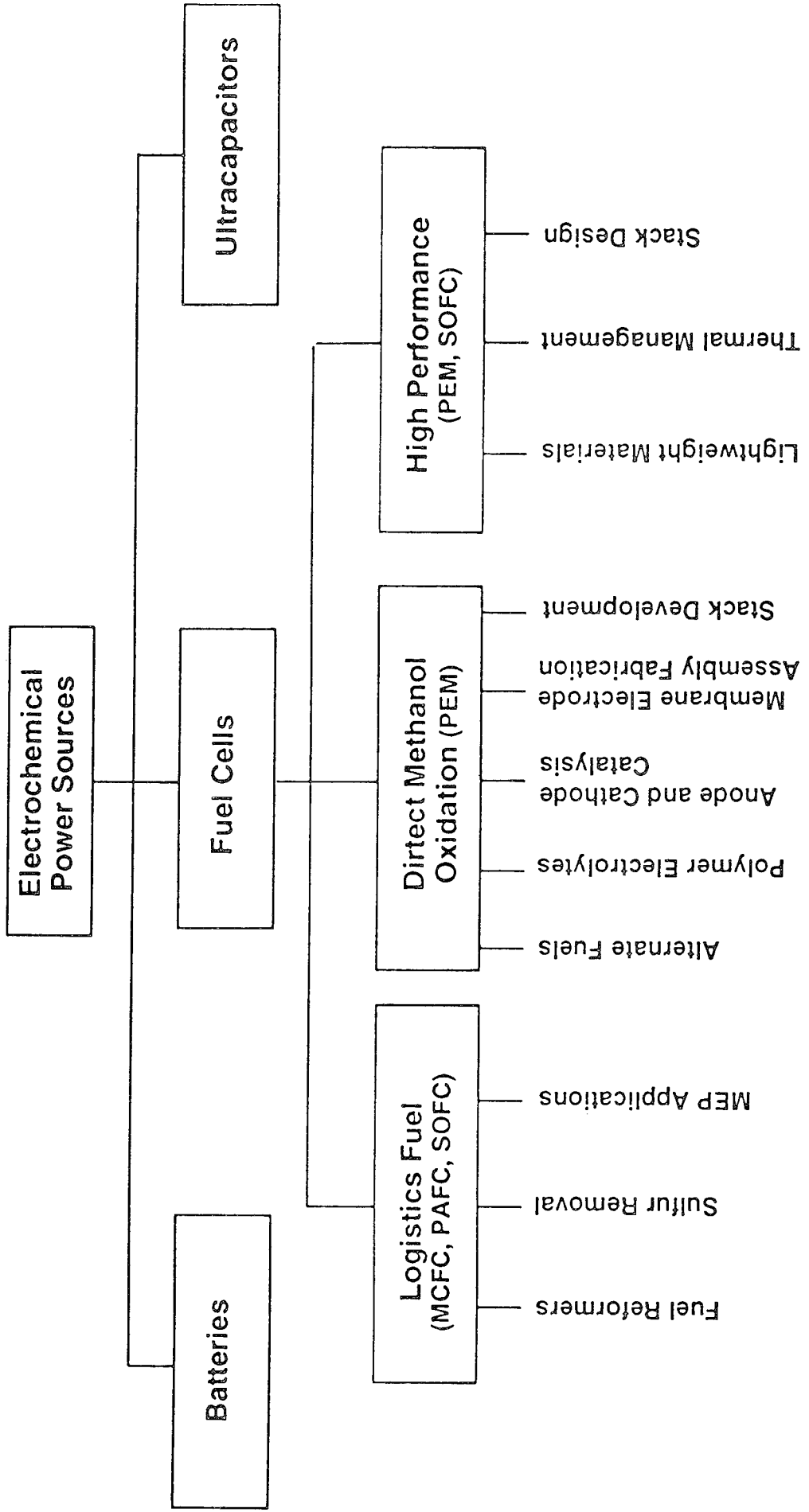
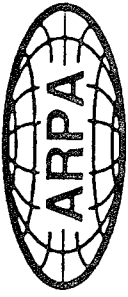
**3701 N. Fairfax Dr.
Arlington, VA 22203-1714**

**phone: (703) 696-2283
fax: (703) 696-2201
email: ldubois@arpa.mil**





ELECTROCHEMICAL POWER SOURCES PROGRAM

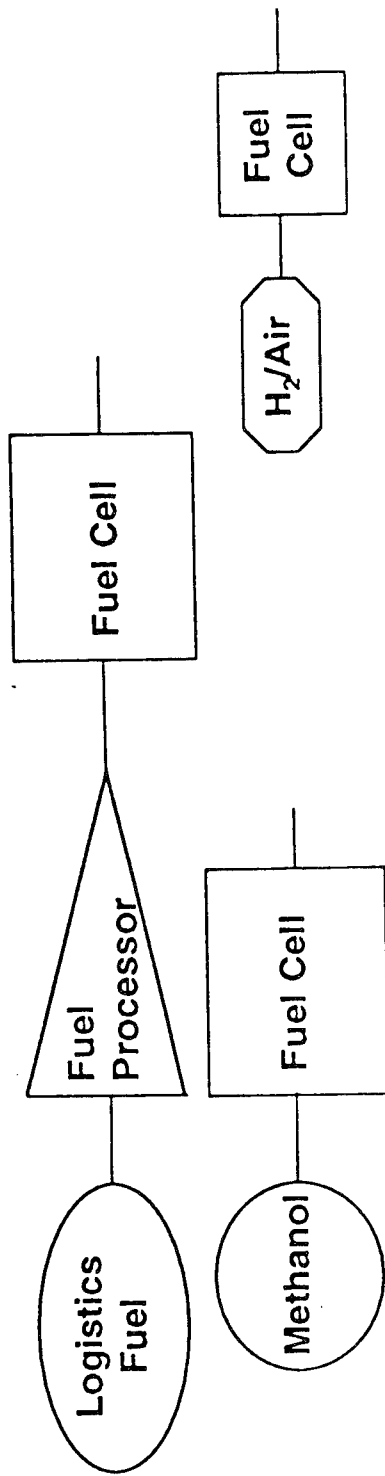




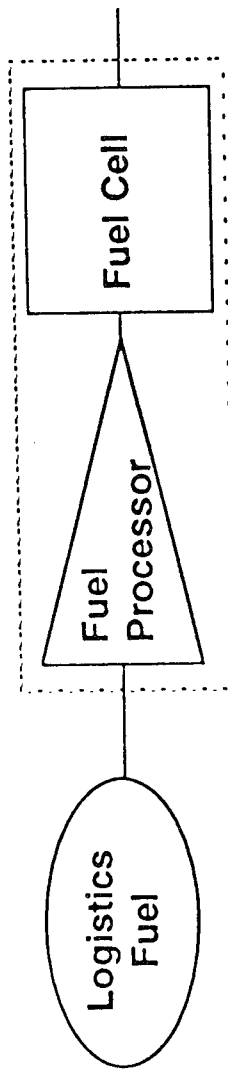
ARPA/DSO FUEL CELL PROGRAM



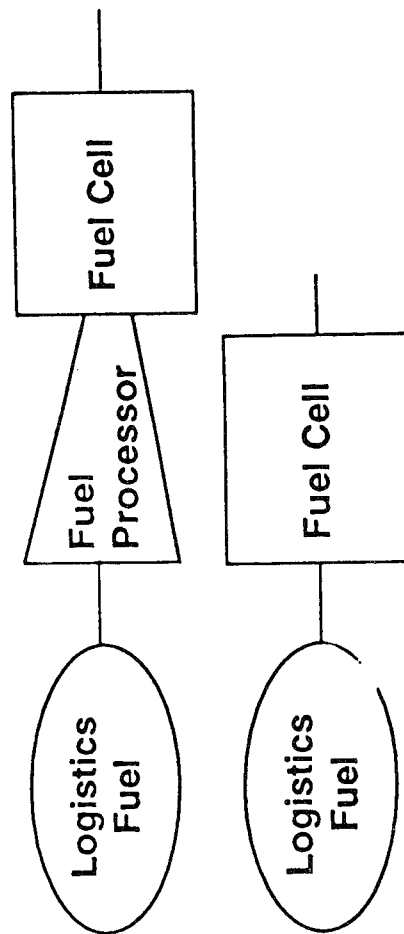
Current



FY 96-98

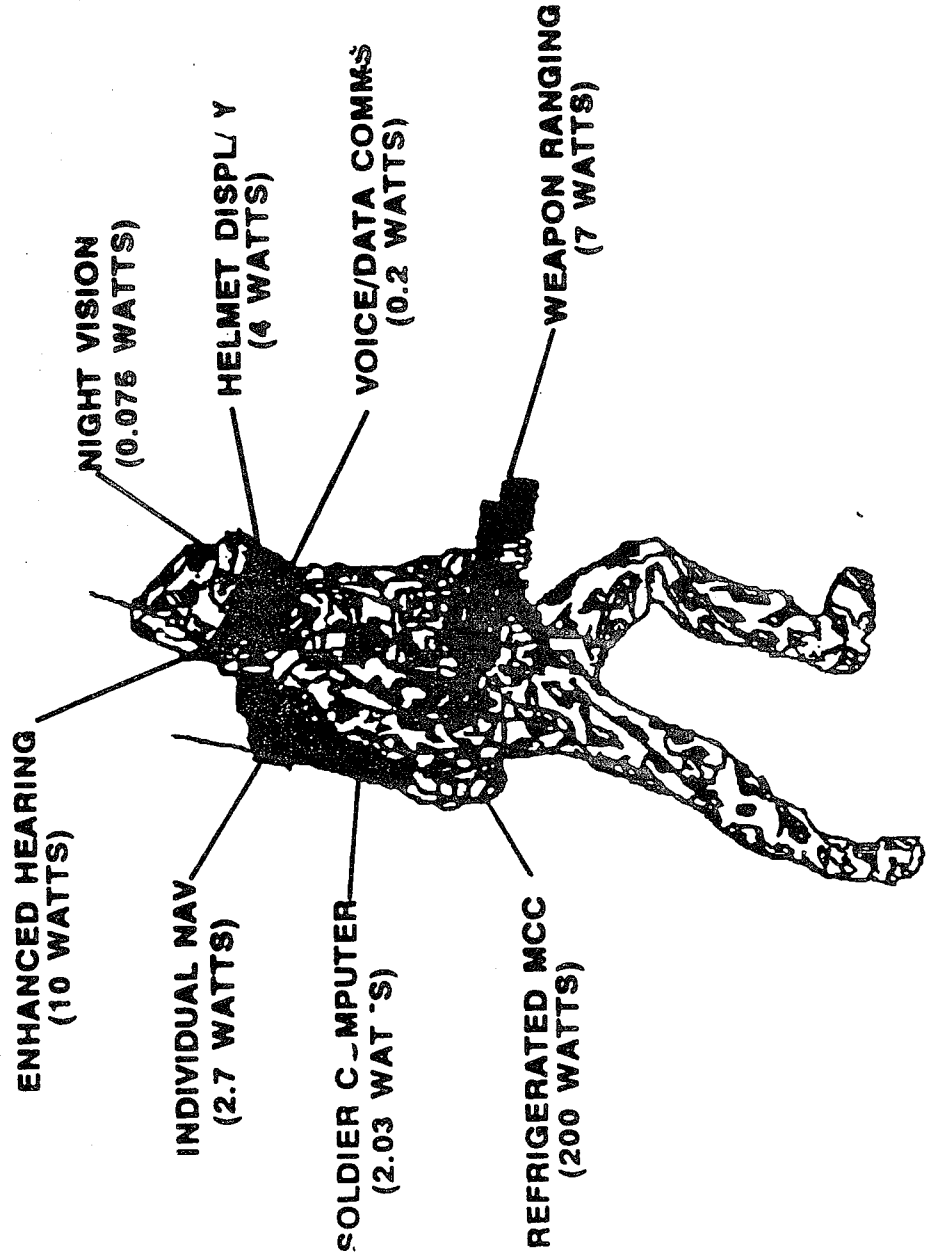


Goal



THE SOLDIER SYSTEM

ANTICIPATED POWER LOADS





Characteristics of Various Rechargeable Batteries



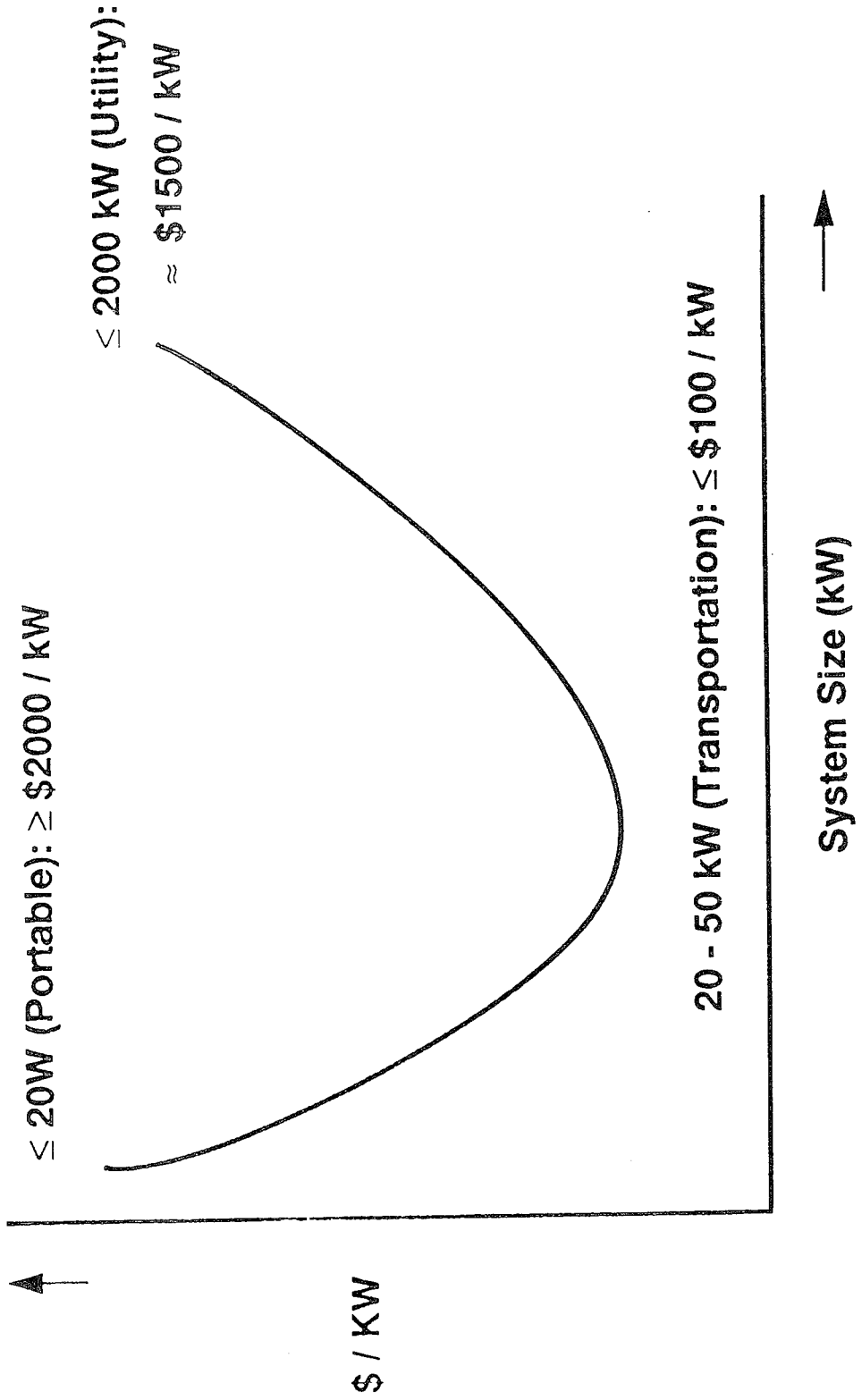
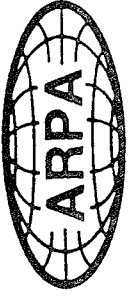
	Lithium Polymer		NiCd	Lead Acid Tabular	Alkaline Non- rechargeable
	Achieved	Development/goal			
Energy Density (Wh/kg) (Wh/l)	300/150* 600/300*	300 600	40 90	35 100	95 210
Power Density (W/kg) (W/l)	700**/70 1400**/140	700**/300 1400**/600	700 1575	70 210	40 88
Voltage (V)	3	3	1.2	2.2	1.5
Self Discharge (%/month)	0.1	0.1	20-50	10-20	0.5
Number of Cycles	>200	>1000	1000	400-600	none
Operating Temperature (°C)	-20 to +70	-40 to +125	-40 to +50	0-100	-20 to +55
Safety	OK	OK	OK	OK	OK
Environment	OK	OK	Cd	Pb	OK
Memory Effect	none	none	yes	none	none
Cost (US\$/kWh) (commercial mass production)	--	400	1500	250	400

* Energy density after 20 cycles when energy density stabilizes at about 50% of initial value.

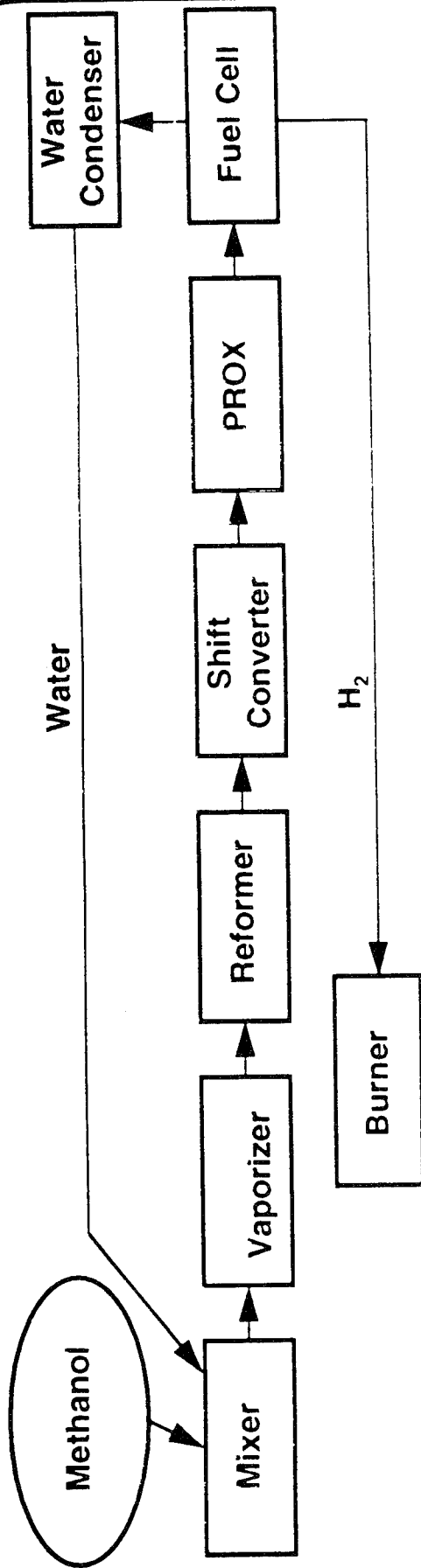
** Power density during short periods of up to 15 seconds.



Approximate Market Entry Price for Fuel Cells

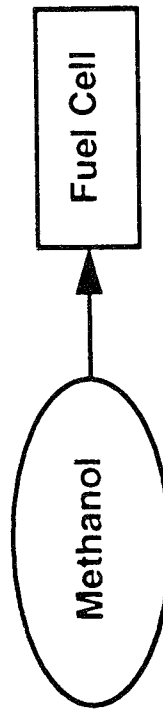


Conventional Approach



- Reduced complexity by factor 4-6
 - Reduced weight and volume by 25-50%
 - Improved reliability and lower maintenance
 - Rapid start capability, rapid transient response
 - Reduced capital and operating costs
 - Lower thermal signature
- => Increased applicability of fuel cells to additional DoD missions

Direct Oxidation

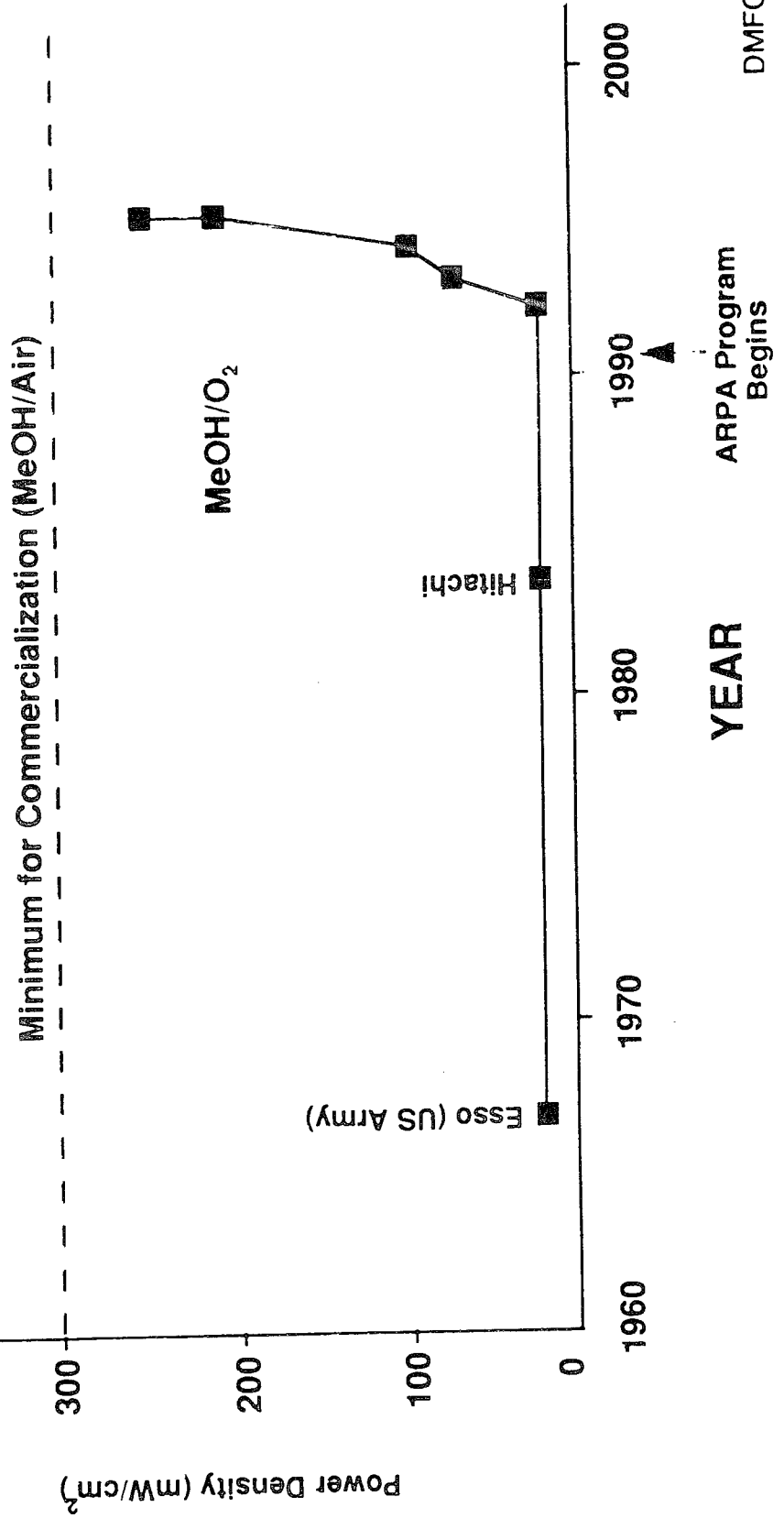




PROGRESS IN DIRECT METHANOL OXIDATION PEM FUEL CELLS

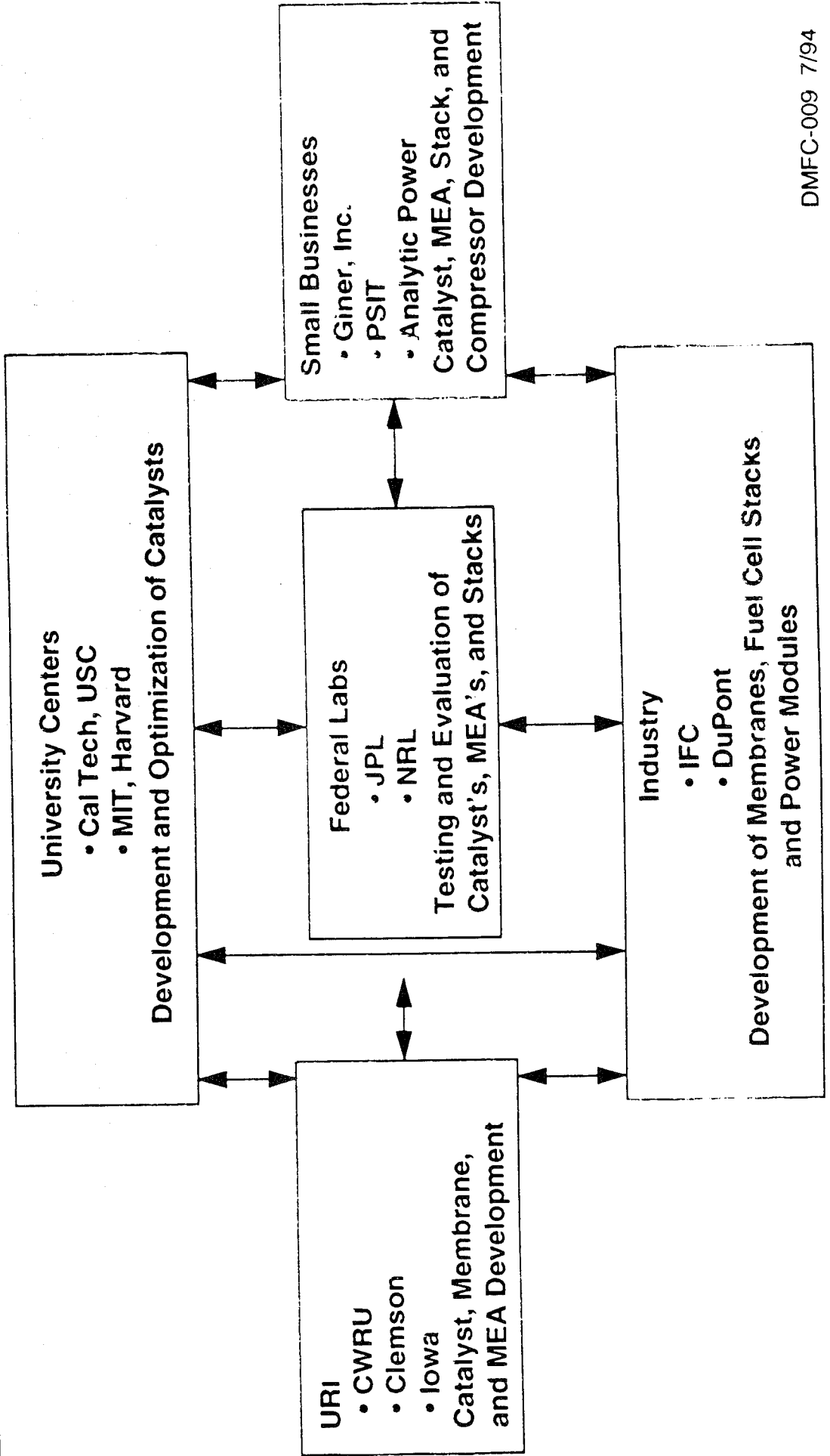


H₂/Air: 600-700 mW/cm²





ARPA Direct Methanol Fuel Cell Program Team Approach





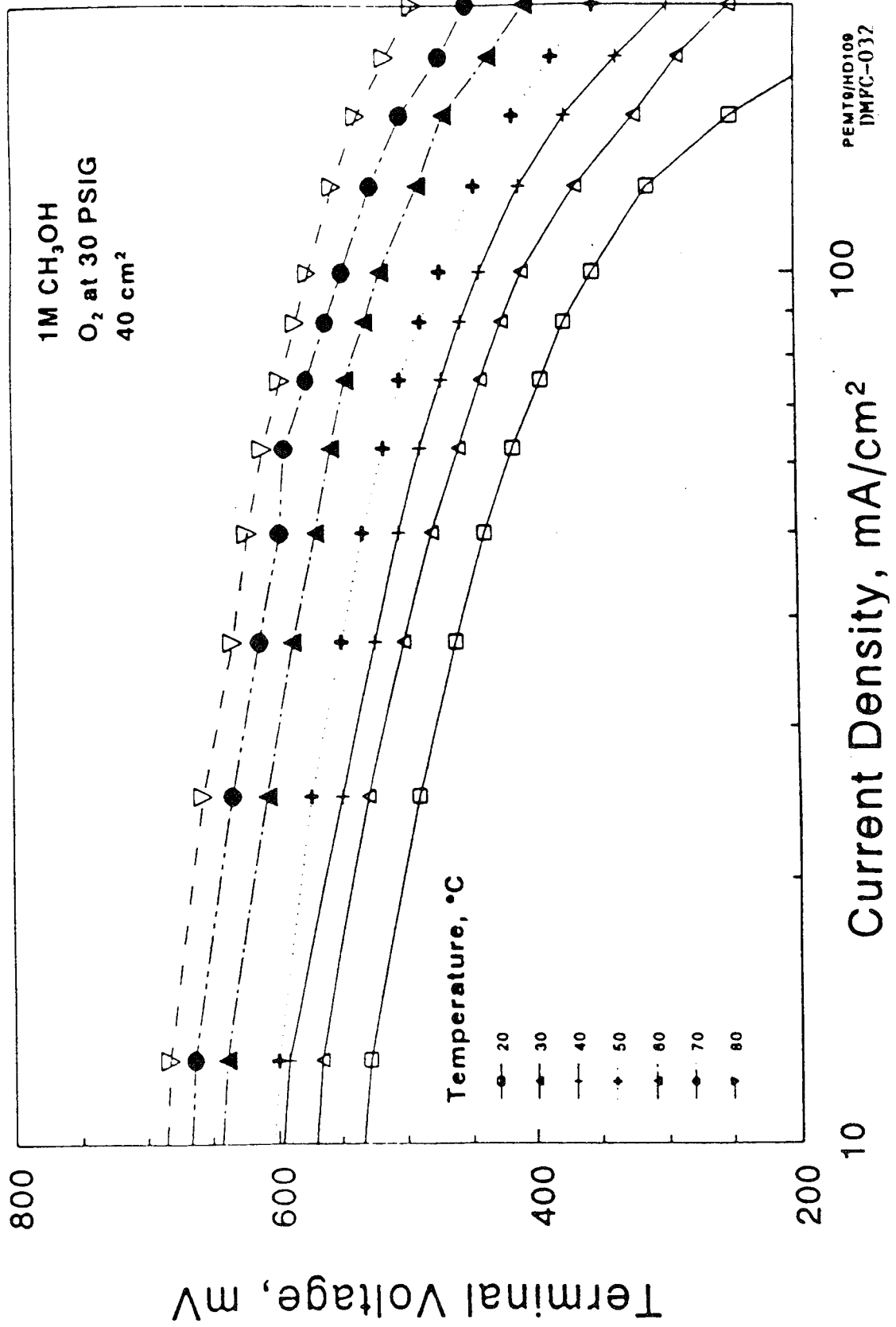
ARPA Direct Methanol Fuel Cell Program Highlights



- Performance - single cell: 0.33 V @ 800 mA/cm² (260 mW/cm²)
0.41 V @ 640 mA/cm² (260 mW/cm² x
258 cm² = 67 W/cell)
- 4 cell stack: 0.40 V @ 430 mA/cm² (172 mW/cm² x
258 cm² => 178 W)
- Efficiency - 25-30% @ <300 mA/cm²
- Fuel Utilization - >70% @ 300 mA/cm²
- Life test - single cell: 0.45 V @ 300 mA/cm² for >200 hrs continuous
(>500 hrs intermittent)
- 5 cell stack: 0.56 V @ 100 mA/cm² (48 W) for ~400 hrs
- Start-up from ambient temperature (20 °C)

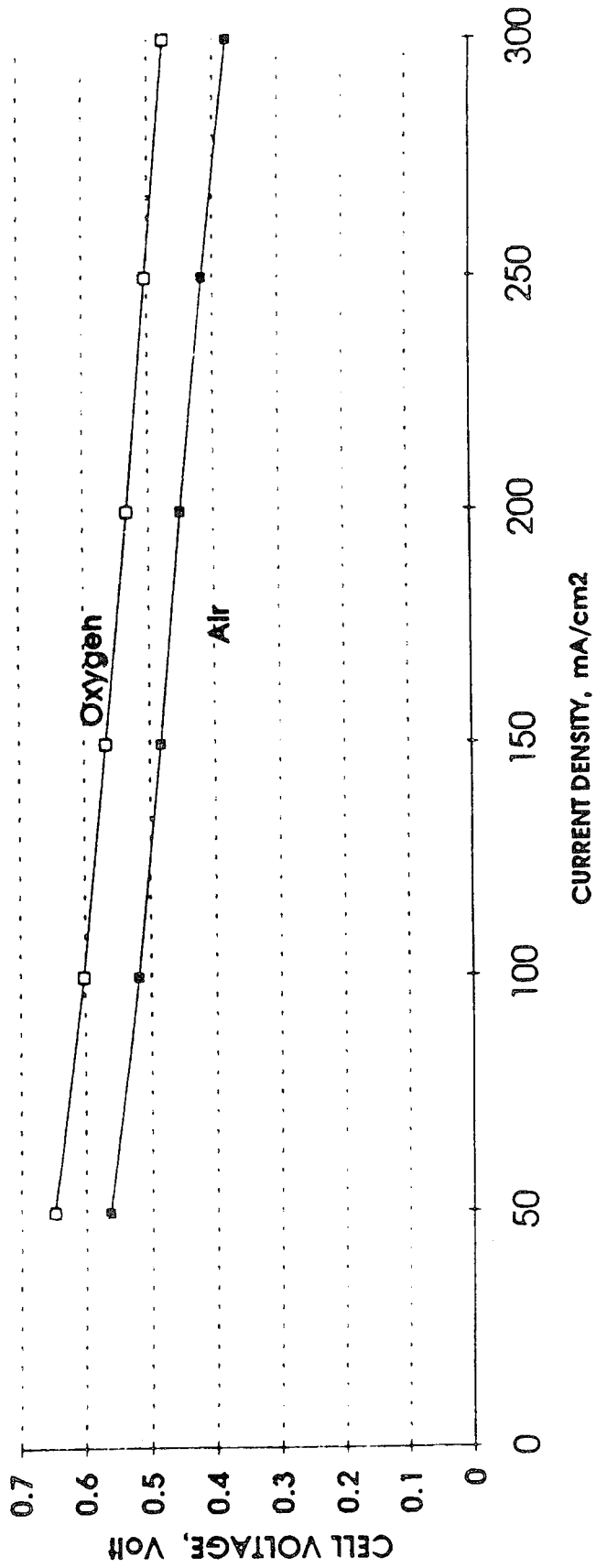
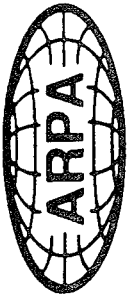


Effect of Temperature on Performance of DMFC

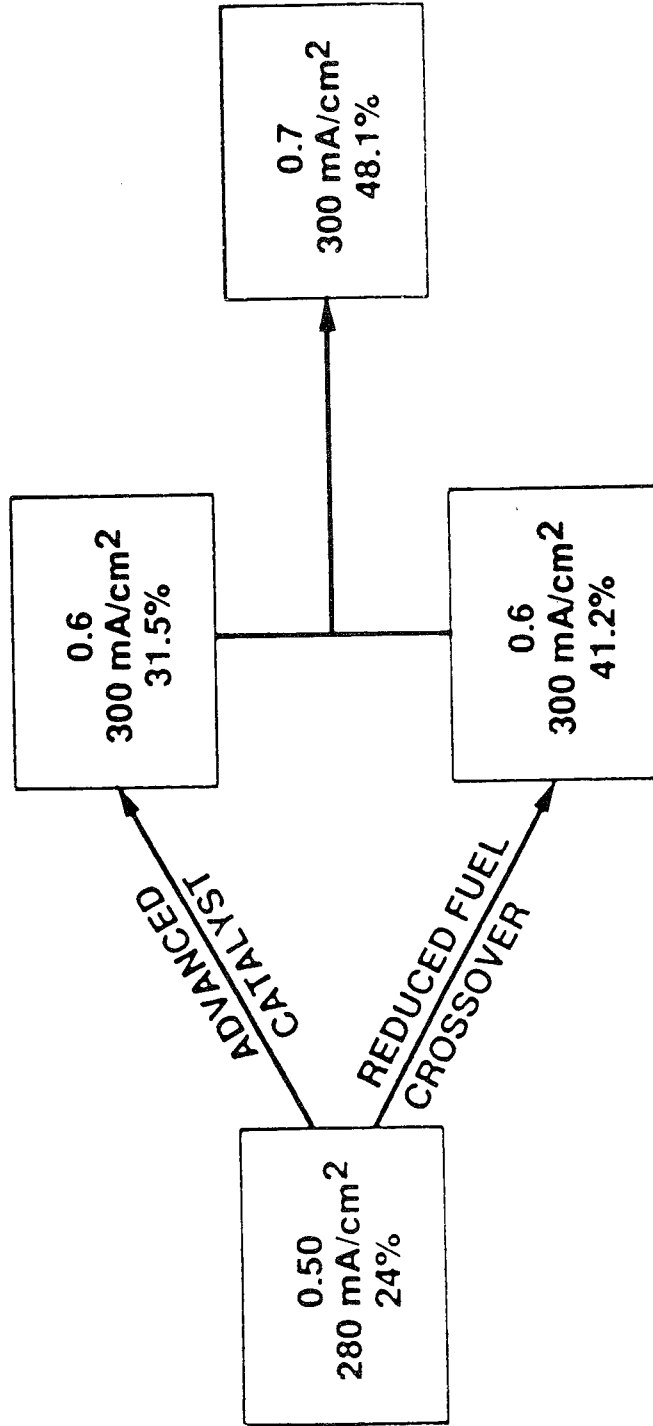




PERFORMANCE OF DIRECT METHANOL FUEL CELL WITH OXYGEN AND AIR



PROJECTED ADVANCES IN DMFC PERFORMANCE



- REDUCING CROSSOVER AND IMPROVING ANODE PERFORMANCE REQUIRED TO ACHIEVE EFFICIENCIES OF ~48%

ENERGY STORAGE SYSTEMS GROUP



Why Methanol?

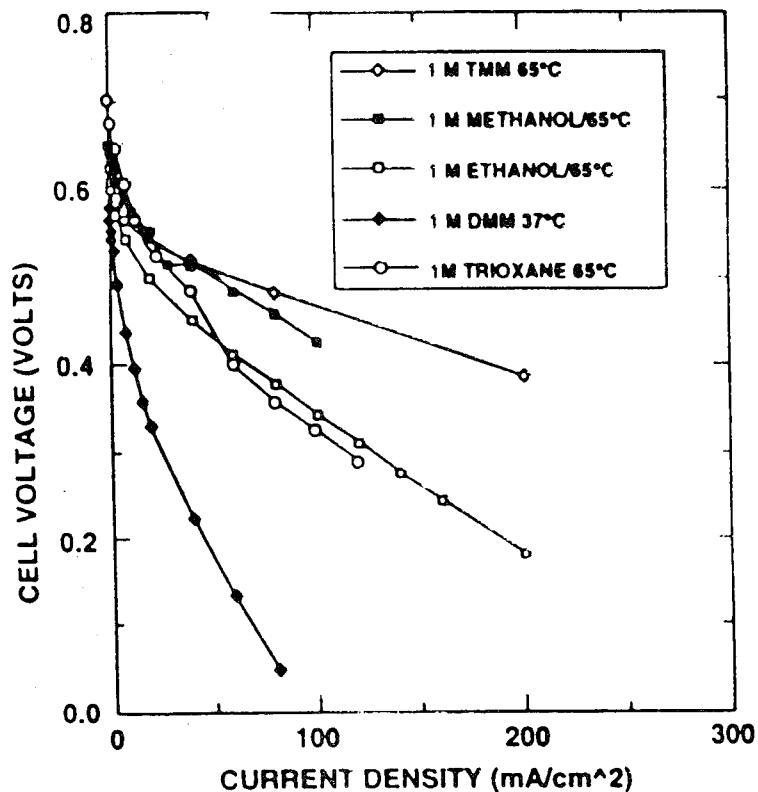


- **Environmentally friendly.**
- **Low vapor pressure liquid fuel.**
- **Ease of handling and distribution.**
- **High power density.**
- **Safe!**
- **Low cost and readily available domestic fuel
(from coal or biomass).**

Properties of Oxygenated Fuels

FUELS	B.P	E°	Ah/g	Wh/KG
HCHO	21°C	1.350	3.57	4820
CH ₃ OH	65 °C	1.21	5.03	6086
C ₂ H ₅ OH	78.5 °C	1.143	6.99	7989
(CH ₃ O) ₂ CH ₂	47 °C	-	5.642*	6826/ 7616@
(CH ₃ O) ₃ CH	100 °C	-	5.056*	6110/ 6825@

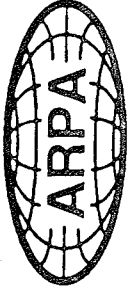
PERFORMANCE OF ALTERNATIVE FUELS



• TRIMETHOXYMETHANE IS AN ATTRACTIVE ALTERNATE TO METHANOL



ARPA Advanced Fuel Cell Technologies Conclusions



- The performance of direct methanol oxidation fuel cells have improved >10 fold in the ARPA program, but further improvements of at least a factor of two are required before commercialization is possible. Technical challenges include inhibiting methanol cross-over, increasing catalyst utilization, and stack scale-up.
- Thermally integrated fuel processor/fuel cell power systems operating on military logistics fuel are possible. Cost and complexity may preclude their early use in the field. Direct oxidation at elevated temperatures has also been demonstrated and may provide a reasonable alternative.
- High performance and high efficiency operation will be required for all fuel cells used in portable and vehicular applications. Operation of PEM and SOFC fuel cell stacks on H₂/air mixtures at greater than 1 kW/kg and 1 kW/liter is possible.

TECHNOLOGY UPDATE I

TECHNOLOGY UPDATE SESSION I

Chairman: Dr. M. Frank Rose (AUSPI)

This session began with introductory remarks from the session chairman reiterating the purpose of the technology update sessions. He requested that each speaker to do several things during the three sessions:

- **Identify the state-of-the-art (SOA)** in the near term,
- **Highlight what is in the laboratory now,**
- Clearly state **what the speaker thinks need be done to advance the SOA**, in small, portable fuel cell technology programs, and
- **Identify key technological issues** to be addressed later in the Working Group discussions.

This session was organized to highlight the SOA in many types of fuel cell programs, as well as in competing battery technologies at the Army Research Laboratory (ARL). The lectures presented included the following:

- "Status of ARL Battery Programs," Dr. Bob Hamlen, Army Research Lab
- "Proton Exchange Membrane Fuel Cells - Basic Research to Technology Development," Dr. Supramaniam Srinivasan, Texas A&M
- "Fuel Cell Development Program for the Army," Mr. Dave Bloomfield, Analytic Power
- "Overview of PEM Fuel Cell Research at ARL," Dr. Michael Binder, Army Research Lab
- "Aluminum/Air Semi-Fuel Cells for Portable Power Applications," Dr. Bhaskara Rao, Explorex

The remainder is a compilation of the technical material as submitted to the Workshop organizers.

PORTABLE POWER SOURCES

R. P. Hamlen
Power Sources Division,
Army Research Laboratory

The key areas of interest in the area of portable power sources are:

- Primary Batteries
- Rechargeable Batteries
- Reserve/Fuze Batteries
- Pulse Batteries and Capacitors
- Portable Fuel Cells
- Thermophotovoltaic Power Generation

With regard to primary batteries, there are two main interests: (a) the next generation primary battery to follow on after the depletion of the present stock of Li/SO₂ batteries, and (b) a battery with the maximum energy density for limited-use applications such as the microclimate cooling unit for the Soldier System.

Rechargeable batteries are becoming relatively more important, both for Special Operations use and as a means of reducing training costs. However, higher energy densities are necessary to avoid the need to carry space batteries during an 8-hour training mission.

Improved lithium-based fuze batteries are required to remove the need for lead-based systems and to enable longer operating times.

High-rate pulse batteries and capacitors are needed for electrothermal gun applications, and for heavy engine starting and for electric vehicle acceleration. Recent advances have been made at ARL in electrochemical capacitors for the latter two applications.

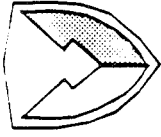
Work is in progress on improving membranes and electrocatalysts for PEM fuel cells, in order to provide for better light-weight fuel cell stacks.

Thermophotovoltaic power generation is being developed as a means of providing silent power for battery charging and other field applications.



US ARMY
RESEARCH LABORATORY

13 Sep 94



ARMY
MATERIEL COMMAND

PORTABLE POWER SOURCES

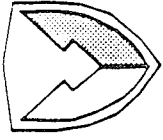
DR R.P. HAMLEN
Director, Power Sources Division
Electronics and Power Sources Directorate

UNCLASSIFIED



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BATTERY RELATED FUNCTIONS

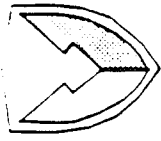


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-
- CONDUCT R&D ON FUTURE BATTERIES
 - DEVELOP IN CONJUNCTION WITH CONTRACTORS
 - APPROVE NEW TYPES FOR USE
 - QC TESTING ON LITHIUM BATTERIES



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**POWER SOURCES
DIVISION**
R.P. Hamlen

**ENERGY
SCIENCES**
S. Gilman

**BASIC RESEARCH
ON:**

- Manportable
 - Batteries
 - Fuel Cells
- Batteries & Capacitors for Electric Weapons

**RESERVE BATTERY
TECHNOLOGY**
A. Goldberg

**Reserve Batteries
for**

- Smart Weapons
- Standby Uses
- Integral Active (Lithium) (Thermal) (Aqueous)

**BATTERY DEV/
ENGINEERING**
H.A. Christopher

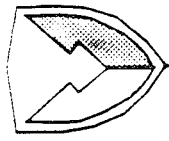
Application Eng.

- Lithium Battery
 - safety
 - producibility
 - analyses
- Battery Product Design & Dev.
- Thermophotovoltaic Systems Dev.



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POWER SOURCES PROGRAMS



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THROWAWAY BATTERIES

- NEXT GENERATION PRIMARY
- SOLDIER SYSTEM PRIMARY

PULSE BATTERIES AND CAPACITORS

- ELECTRIC GUN
- ELECTRIC DRIVE VEHICLE

RECHARGEABLE BATTERIES

- IMPROVED ENERGY DENSITY
- LOW COST

FUEL CELLS

- SOLDIER SYSTEMS
- TANK SILENT WATCH
- FIELD POWER GENERATION

RESERVE BATTERIES

- LI-BASED SYSTEMS
- LONGER MISSION LIFE

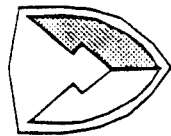
ALTERNATIVE POWER SOURCES

- THERMOPHOTOVOLTAIC ENERGY
GENERATION



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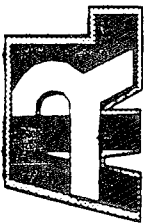
POWER SOURCES DIVISION



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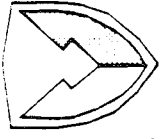
KEY BATTERY CONCERNS

- I. PERFORMANCE
 - ENERGY, POWER, WEIGHT, TEMPERATURE
 - SAFETY-TRANSPORTATION, USE, DISPOSAL
 - SMART CHARGER, STATE-OF-CHARGE
- II. COST
 - PEACETIME TRAINING
- III. PROLIFERATION OF BATTERY TYPES
 - PRIOR TO LITHIUM-LITTLE CONTROL
 - NOW - STANDARD FAMILY



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KEY BATTERY CONCERNS

(Continued)

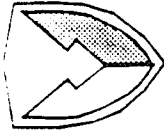
- IV. DIVERSIFIED APPLICATIONS
 - COMMUNICATIONS-ELECTRONICS
 - TANK STARTING AND SILENT WATCH
 - PULSE POWER
 - AIRCRAFT STARTING

- V. INDUSTRIAL BASE

FAMILY OF STANDARD BATTERIES



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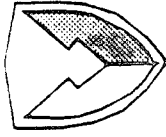
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CAPACITY CURRENT VOLTAGE	NON RECHARGEABLE LOW POWER Li/SO ₂	RECHARGEABLE LOW POWER Ni-Cd	NON RECHARGEABLE HIGH POWER Li/SO ₂	RECHARGEABLE HIGH POWER Ni-Cd
3	BA-5567/U			
6	BA-5372/U		BA-5847/U	BB-X847/U
12	BA-5588/U	BB-588/U		
24			BA-5590/U	BB-590/U

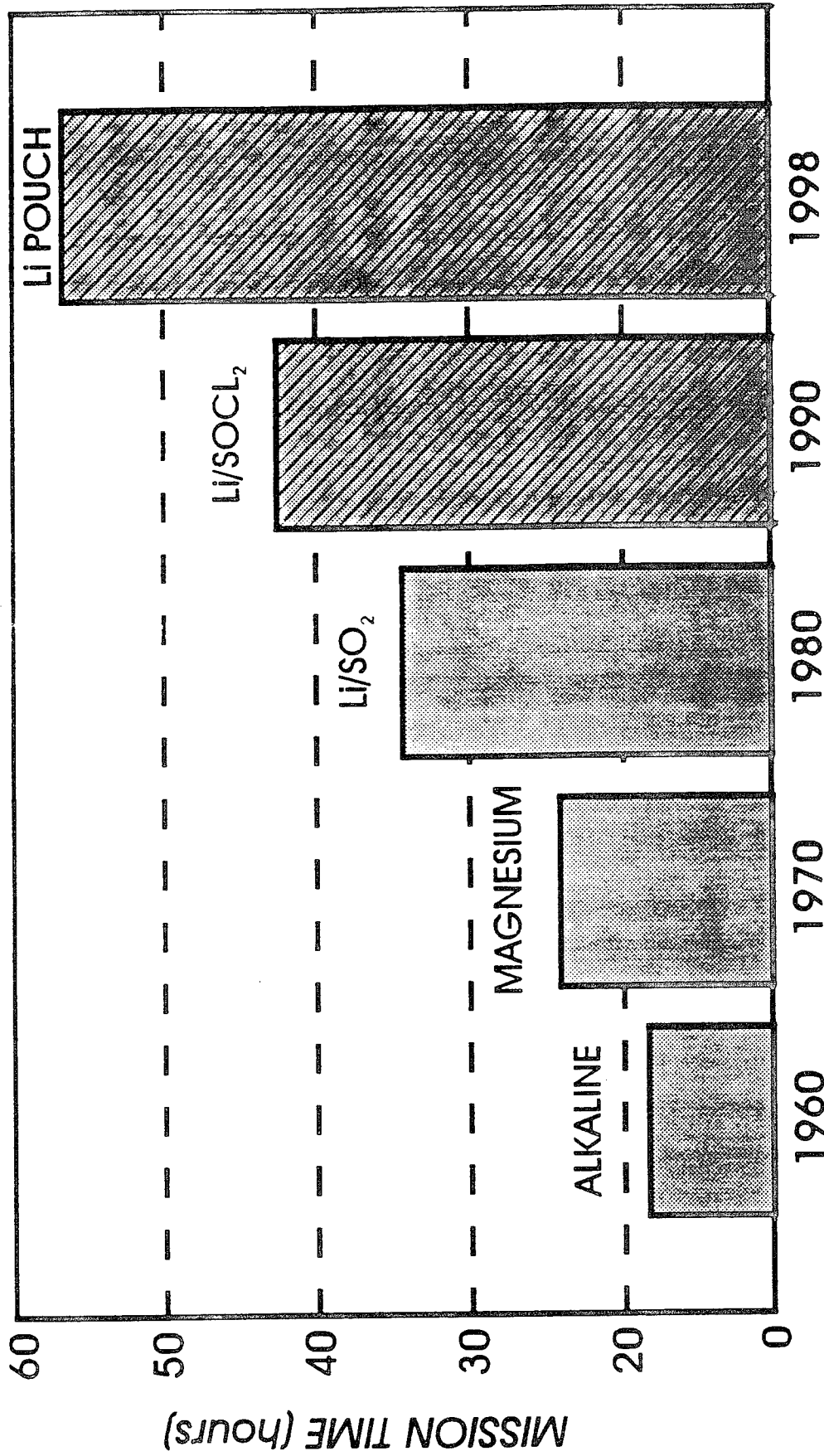


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ADVANCES IN PRIMARY BATTERIES FOR HIGH POWER DENSITY APPLICATIONS*



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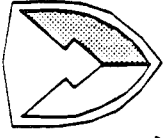


* SPECIFIC DATA FOR THE BA-5590/J IN THE SINGGARS RADIO



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LITHIUM-MANGANESE DIOXIDE PRIMARY BATTERIES



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CYLINDRICAL CELLS

- SMALL CELLS- AVAILABLE: CAMERAS, SMOKE ALARMS
- D CELLS - AVAILABLE: SEARCH AND RESCUE BEACONS (8 Ahrs.)

POUCH CELLS

- MAXIMIZE USE OF AVAILABLE BATTERY VOLUME
- MORE ENERGY PER UNIT OF VOLUME
- EASIER/CHEAPER TO MANUFACTURE THAN SPIRAL WOUND CELLS
- FOIL CONSTRUCTION - NO SHRAPNEL
- POUCH CELL FOR BA-XX90/U 14 Ahrs.

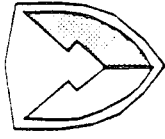
TECHNICAL CONCERNS

SAFETY, LOW TEMP OPERATION, SHELF LIFE



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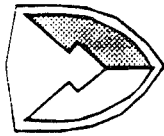
RECHARGEABLE BATTERIES

- TRAINING
- SOF
- TANK STARTING/SILENT WATCH
- AIRCRAFT STARTING



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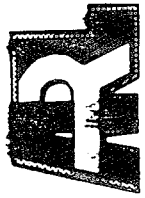
ARMY
MATERIEL COMMAND

SINCGARS ALTERNATIVES AVAILABLE TODAY

BA-5590/U BB-590/U BB-490/U
LITHIUM S02 (P)* NICKEL-CADMIUM (R)** LEAD-ACID (R)

BATT/MISSION (8 hrs)	1	2	2
MISSIONS/BATT	3.1	0.79	0.55
WT/MISSION	2.2 LBS	7 LBS	6.8 LBS
COST/YR	\$4,393	\$496	\$2675
DISPOSAL/YR	\$238	\$3	\$0

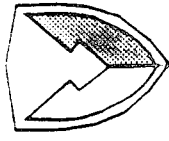
* (P) PRIMARY
** (R) RECHARGEABLE



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COMPARISON OF EXISTING AND FUTURE RECHARGEABLE BATTERIES

BB590/U SIZE, 2.45"x5"x4.4"-24V.



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PRODUCTS AVAILABLE

Wh/Kg AH SAFETY CYCLES COMMENTS/LIMITATION

Ni Metal Hydride	42	3.4	+	200+	Potential For Improvement
NiCd	30	2.0	+	250+	Heavy/Low Mission Life
Lead-Acid	25	1.6	+	100	Heavy/Lower Mission Life



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COMPARISON OF EXISTING AND FUTURE RECHARGEABLE BATTERIES

BB590/U SIZE, 2.45"x5"x4.4"-24V.

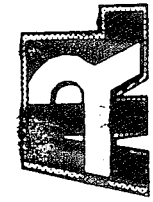


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Continued

<u>FUTURE CANDIDATES</u>	<u>Wh/Kg</u>	<u>AH</u>	<u>SAFETY</u>	<u>CYCLES</u>	<u>COMMENTS/LIMITATION</u>
Li-ion	88	4.5	?	200+	Safety in Large Sizes
Metallic Li	110	5.0	?	50	Safety/Risk
Metallic Li/Polymer	150	7.0	?	50	Safety/Risk TBD
Rech. Alkaline	60	4.5*	+	10-30	Power/Temp. Limited

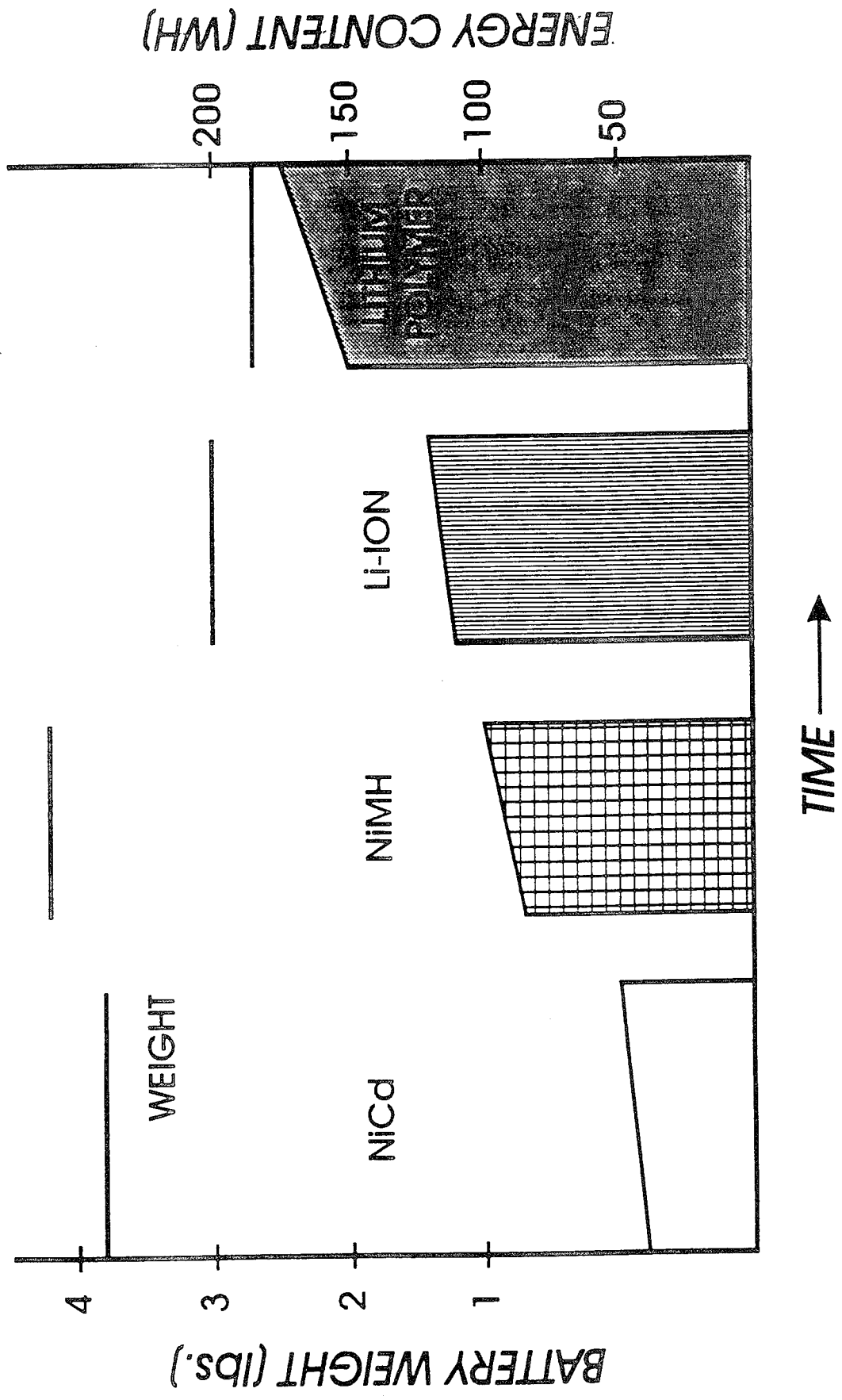
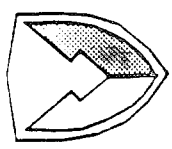
* Low Rates Only



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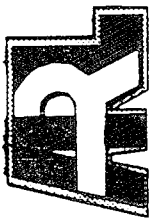
RECHARGEABLE BB590/U TYPE BATTERY FOR SOF C-E APPLICATIONS

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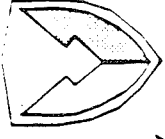
ELECTROCHEMICAL RESEARCH

GOAL	IMPACT/APPLICATIONS
<p>RECHARGEABLE LITHIUM BATTERIES</p>	<ul style="list-style-type: none"> - 1/10 cost of throwaway battery - Over 2 1/2x energy density of secondary battery - Reduced logistics if primary charging source available
<p>ELECTROCHEM CAPACITORS</p>	<ul style="list-style-type: none"> - Hybrid or electric vehicles: acceleration, dynamic braking - Conventional vehicles: engine starting
<p>PULSE POWER BATTERIES</p>	<ul style="list-style-type: none"> - Electric guns - Hybrid or electric vehicles
<p>FUEL CELLS</p>	<ul style="list-style-type: none"> - Manpack power source for long-duration microclimate cooling - Power Source for special operations - Stealth vehicles



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RECHARGEABLE LITHIUM BATTERIES



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BASELINE TECHNOLOGY

- Ni/Cd (ENERGY DENSITY = 40 Whr/kg)
- Ni/METAL HYDRIDE (ENERGY DENSITY = 60 Whr/kg)

GOAL

- MATERIALS AND BASIC TECHNOLOGY FOR:
 - ENERGY DENSITY > 100 Whr/kg, ALL-TEMP OPERATION
 - DUAL USE, HIGHEST USER SAFETY

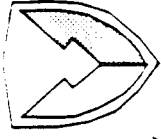
APPROACH

- IMPROVED LIQUID ELECTROLYTES FOR "Li-Ion"
(I.E. LiC₆ ANODES) BATTERIES
- IONICALLY CONDUCTING POLYMER ELECTROLYTES
FOR SOLID-STATE BATTERIES



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RECHARGEABLE LITHIUM BATTERIES



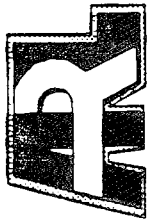
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ACCOMPLISHMENTS

- HIGH CONDUCTIVITY LIQUID ELECTROLYTES AVAILABLE FOR "Li-Ion" BATTERIES
- NEW POLYMER ELECTROLYTES FORMULATED

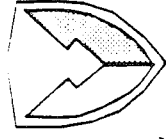
FUTURE WORK

- LIQUID ELECTROLYTES: STABILITY WITH Li_xC_6
- POLYMER ELECTROLYTES: FORMULATION/ DEMONSTRATION OF PROTOTYPE CELLS
- EXPLORATION OF CARBON FIBER CATHODES



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POWER MANAGEMENT



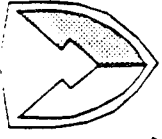
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- **SIGNIFICANT POWER/ENERGY SAVINGS CAN BE ACHIEVED IN ELECTRONIC EQUIPMENT BY:**
 - Applying emerging device and architectural technologies
 - Low voltage analog/digital microelectronics
 - Reconfigurable parallel and pipelined architectures
 - Employing judicious engineering principles throughout the requirements/design/development process
 - Automatic shutdown, adaptive "sleep" modes, etc.



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POWER MANAGEMENT



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- CUMULATIVE EFFECT: POWER SAVING RANGING FROM 10% TO 90%

DEPENDING UPON:

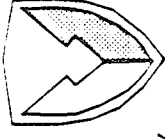
- Willingness to incur moderate increases in NRE and production costs
- System-Specific Performance and Operational Use

- ACQUISITION COST INCREASES CAN BE EASILY RECOVERED BY SAVINGS IN BATTERY COSTS WITHIN 1 TO 3 YEARS



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POWER SOURCES DIVISION



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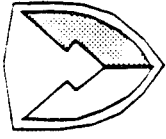
KEY OBJECTIVES FOR FUTURE

- IMPROVED PRIMARY BATTERIES FOR SOLDIER SYSTEMS, NEXT GENERATION C4I THROWAWAY, LASER, RADAR AND NV SYSTEMS
- IMPROVED NEAR-TERM (NiMH) AND LONG-TERM (Li-ION) RECHARGEABLE BATTERIES (C4I, SOLDIER SYSTEM, TANK)
- RESERVE/ACTIVE BATTERIES FOR LONGER-LIVED, SMART MUNITIONS



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POWER SOURCES DIVISION



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KEY OBJECTIVES FOR FUTURE

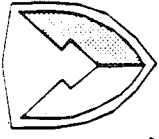
Continued

- HIGH RATE BATTERIES AND CAPACITORS FOR ELECTRIC WEAPONS AND VEHICLES
- FUEL CELLS FOR LONG-LIFE PORTABLE USE, TANK SILENT WATCH
- PORTABLE THERMOPHOTOVOLTAIC POWER GENERATORS



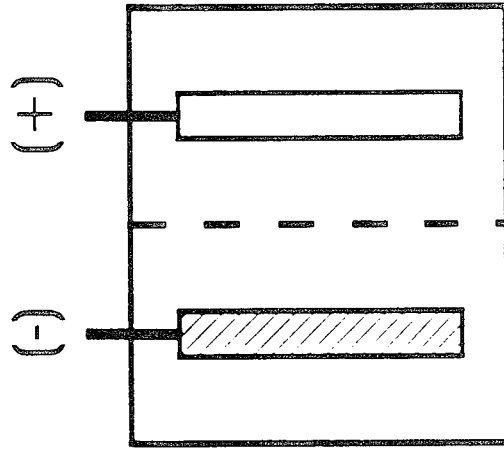
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BATTERIES VS FUEL CELLS

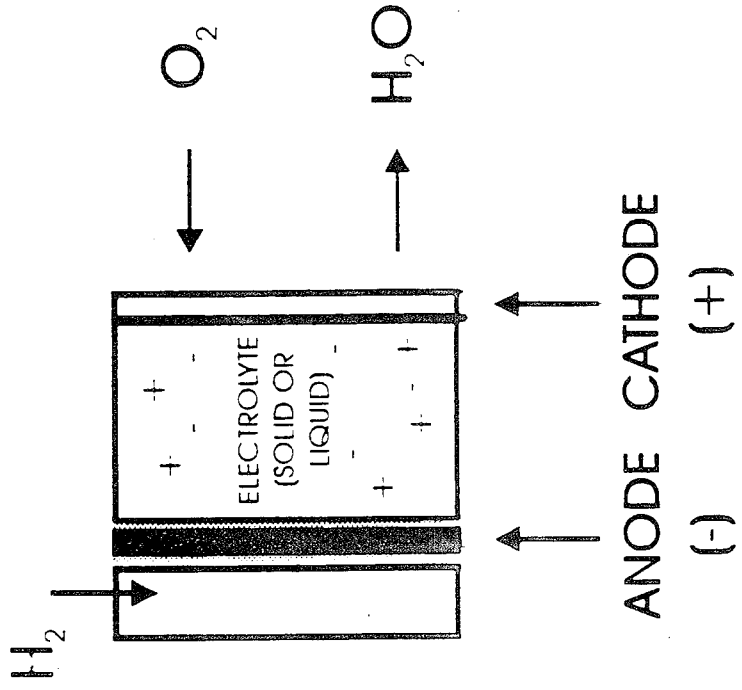


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BATTERY
SELF CONTAINED



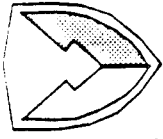
FUEL CELL
CONTINUOUS FEED





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FUEL CELLS



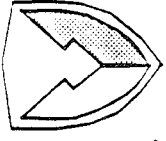
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- BATTERY CHARGER
- LONG-DURATION C4I MISSIONS
- SOLDIER SYSTEMS
- TANK SILENT WATCH
- SILENT VEHICLES



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FUEL CELLS



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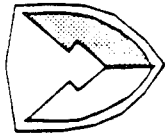
CURRENT PROGRAM

- IMPROVED MEMBRANES FOR POLYMER ELECTROLYTE MEMBRANE (PEM)
- IMPROVED ELECTROCATALYSTS FOR H₂ AND METHANOL FUEL CELLS
- HIGH POWER DENSITY H₂/PEM FUEL CELL STACKS



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THERMOPHOTOVOLTAIC POWER GENERATION



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- NIGHT-TIME USE OF SOLAR PANELS
- MULTI-FUEL CAPABILITY
- SILENT OPERATION
- REMOTE BATTERY CHARGING

EXTENDED ABSTRACT

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

Supramaniam Srinivasan, A. Cesar Ferreira, Renaut Mosdale,
Shinichi Hirano, Seong Min Lee, Felix N. Büchi and A. John Appleby
Center for Electrochemical Systems and Hydrogen Research
Texas Engineering Experiment Station
Texas A&M University System
College Station, Texas 77843-3402

Interest in fuel cell systems for space power, power generation, electric vehicle, portable power, remote power and other applications started in the late 1950s and was further stimulated by the energy crisis of 1973 and by the increasing environmental problems and legislations of the late 1980s and the early 1990s. Due to the relative abundance of natural gas, there is a great stress on it as the primary fuel for fuel cell systems. In the molten carbonate (MCFC) and solid oxide (SOFC) fuel cell systems, this fuel can be used directly with internal reforming in the anode chambers (due to the high grade heat produced because of the irreversible losses in the fuel cell). However, for the low and intermediate temperature fuel cell systems, (proton exchange membrane fuel cell - PEMFC, alkaline fuel cells - AFC, phosphoric acid fuel cells - PAFC), natural gas will have to be converted to high purity hydrogen, which gas will thus be the anodic reactant. Due to the high specific energy and energy density of methanol (about half as that of gasoline or diesel fuel), there is great interest in developing direct methanol fuel cells (DMFCs) for the electric vehicles, portable power and remote power applications. Though the theoretical thermodynamic efficiencies ($\epsilon_r = \Delta G/\Delta H$) for fuel cells are relatively high ($\epsilon_r > 80\%$), the practical efficiencies, during power generation, are considerably less ($\epsilon = 35$ to 50%), because of the slowness of the electrode reactions (activation overpotential), mass transport limitations (concentration overpotential) and electronic/ionic resistances (ohmic overpotential). For the low and intermediate temperature fuel cells (PEMFC, AFC, PAFC and DMFC), activation overpotential is the main cause of efficiency losses, while for the high temperature fuel cells (MCFC and SOFC), mass transport and ohmic overpotentials play a predominant role.

For the portable power applications, the low temperature fuel cells (PEMFC, AFC and DMFC) are the desirable candidates. In order to attain high energy efficiencies and high power densities, which are essential, reduce the weight and volume of the fuel cell power sources for these applications, it is vitally necessary to minimize activation, mass transport and ohmic overpotentials. When using organic fuels (steam-reformed/shift-converted or direct) as the anodic reactant and air as the cathodic reactant, the PEMFCs and PAFCs have clear advantages over the AFCS. Until the present time, the energy efficiencies and power densities in a DMFC have been relatively low because of significant activation overpotentials at both the anode and the cathode, while great strides have been made in the attainment of high level performances in PEMFCs with H_2 and air as reactants.

The R&D activities at Texas A&M University cover the spectrum of basic research to technology development on PEMFCs. The highlights of the accomplishments on these activities are summarized as follows:

- (i) first demonstration of nearly equivalent performances in PEMFCs with ten times lower loading of platinum than in the state-of-the-art PEMFCs (i.e., 0.4 vs. 4.0 mg/cm² of Pt) by Nafion impregnation of the electrode;
- (ii) localization of platinum near front surface electrodes, by using a higher percentage of platinum on carbon (20 or 40% Pt/C rather than 10% Pt/C as in state-of-the-art phosphoric acid or alkaline fuel cells) and by sputter-deposition of a thin layer of platinum on the front surface (0.05 mg/cm²), to minimize mass transport and ohmic resistances in the active layer, which is necessary to attain high current densities;
- (iii) enhancing the utilization of platinum by optimization of the structure and composition of the electrodes - use of microgram levels of Pt and alternate electrocatalysis and using the optimum amount of Teflon in the electrode;
- (iv) optimization of the hot-pressing conditions of the Nafion impregnated electrodes and of the operating conditions - humidification of hydrogen at 10°C higher and of oxygen or air at 5°C higher than the cell temperature and pressurization to attain high energy efficiencies and power densities;
- (v) selection and evaluation of alternate proton conducting membranes and correlating electrochemical performances of PEMFCs with the physico-chemical properties of membranes;
- (vi) development of a micro-electrode technique to determine electrode kinetics of fuel cell reactions, and of mass transport parameters of fuel cell reactants in membranes. Experiments were also conducted in single cells using steady-state and transient techniques to give similar data. The electrode kinetic parameters were shown to be nearly the same for the micro-electrodes as for the non-crystallites of Pt supported on carbon in the fuel cell electrode;
- (vii) an experimental and theoretical mass transport analysis with O₂/He, O₂/Ar, O₂/N₂ gas mixtures as the cathodic reactant to demonstrate that increase in the slope of the E vs. i plot is due to mass transport in the active layer and that the departure from linearity of the E vs. i plot is due to water films or droplets in the diffusion-substrate layer, which thus impedes the transport of the cathodic reactant. The mass transport losses were lower with He than with Ar or N₂ as inert gas. More recently, it was shown that an empirical equation $E = E_0 - b \log i - R_i - m \exp(ni)$ excellently fits the experimental cell potential (E) vs. current density (i) data;
- (viii) experiments to show that there is no net transport of water from the anode to cathode during PEMFC operation;
- (ix) modeling analysis of cooling methods and experimentally showing advantages of the evaporative cooling method which also serves the purpose of humidification, with air - the cathodic reactant;
- (x) engineering design and scale-up leading towards multicell stack developments of PEMFC (1 to 10 kW) with low platinum loading electrodes; and
- (xi) an experimental analysis of PEMFCs illustrating the electrocatalytic and depolarization problems in PEMFCs using methanol directly as the fuel.

PROTON EXCHANGE MEMBRANE FUEL CELLS -
BASIC RESEARCH TO TECHNOLOGY
DEVELOPMENT

Supramaniam Srinivasan*, A. Cesar Ferreira, Renaut Mosdale, Shinichi Hirano,
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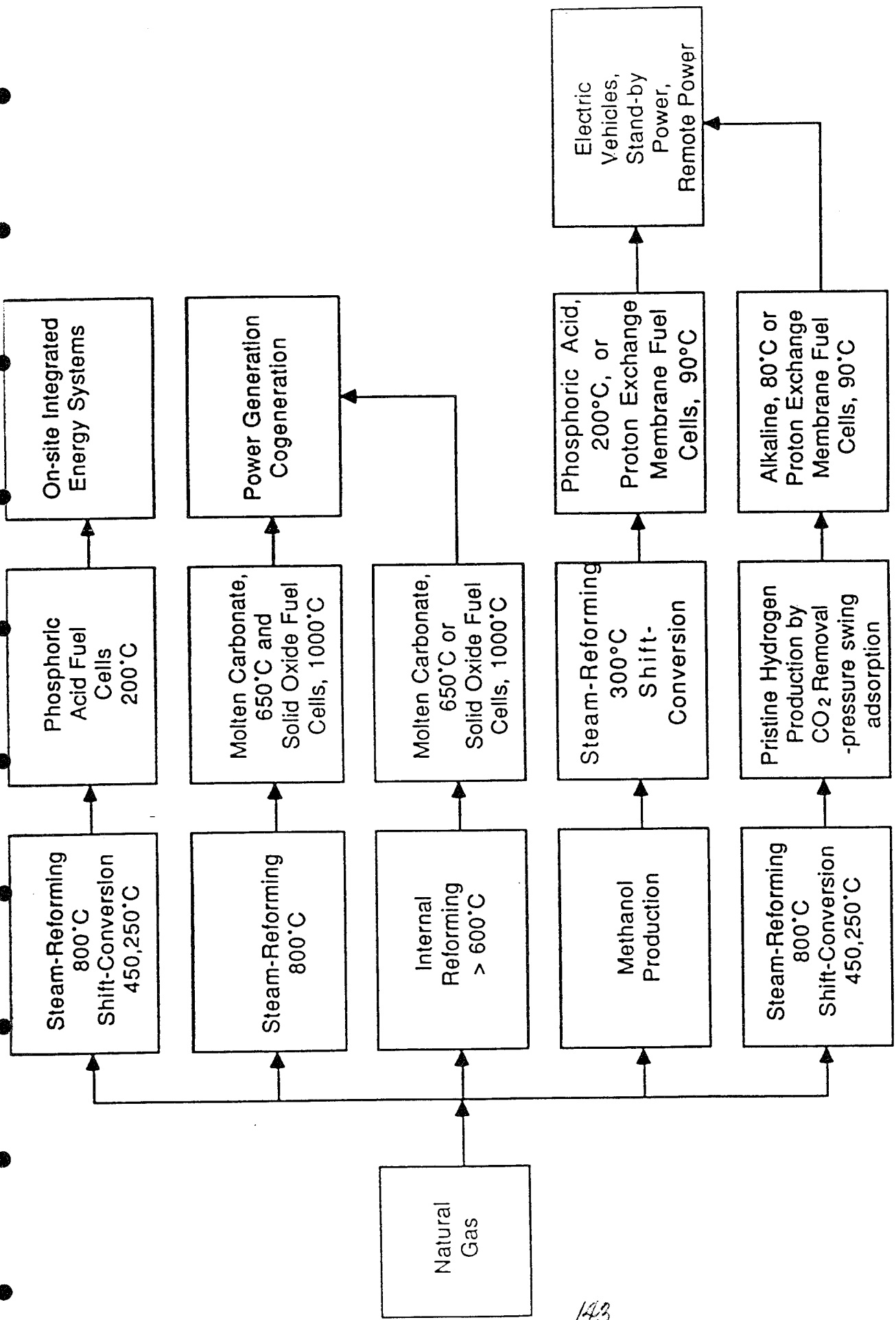
*Presentation at "Prospector VII: Small Fuel Cells for Portable Power" Workshop,
Sponsored by U.S. Army Research Office and Auburn University

Durham, North Carolina
October 31 - November 3, 1994

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

TOPICS

- Rationale for PEMFCs - Alternate FCs, Thermodynamics and Electrode Kinetics
- Optimization of Structures of Electrodes and of Membrane and Electrode Assemblies - Lower Pt Requirement, Attain High Energy Efficiency and Power Densities
- Proton Conducting Membranes - Physico-chemical Characteristics and PEMFC Performance
- Mass Transport - Experimental and Modeling Analysis
- Water Management and Thermal Management
- Multicell Stack Development - with Low Platinum Loading Electrodes
- Conclusions
- Techno-economic Challenges
- Acknowledgements - Sponsors, Former Coworkers



Modes of Utilization of Natural Gas in Fuel Cells and Fuel Cell Applications

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OVERVIEW OF FUEL CELL TECHNOLOGY

BASIC EQUATIONS

• THERMODYNAMICS



$$\Delta G^0 = -56.32 \text{ kcal/mol} \quad (2)$$

$$\Delta H^0 = -68.12 \text{ kcal/mol} \quad (3)$$

$$\Delta G^0 = -n F E_r \quad (4)$$

$$E_r^0 = 1.229 \text{ V} \quad (5)$$

$$E_r = E_r^0 + \left(\frac{\partial E}{\partial T} \right)_P (T - 298) + \frac{\Delta S}{nF} (T - 298) \quad (6)$$

$$E_r = E_r^0 - \frac{\Delta nRT}{nF} \ln P \quad (7)$$

OVERVIEW OF FUEL CELL TECHNOLOGY

BASIC EQUATIONS (Contd.)

- ELECTRODE KINETICS

$$E = E_0 - \eta_a - \eta_{\text{conc}} - \eta_{\text{ohm}} \quad (8)$$

UP TO END OF LINEAR REGION, FOR LOW AND INTERMEDIATE TEMPERATURE FUEL

$$E = E_0 - b \log i - Ri \quad (9)$$

where $E_0 = E_r + b \log i_0$ (10)

$$\frac{dE}{di} = -\frac{b}{i} - R \quad (11)$$

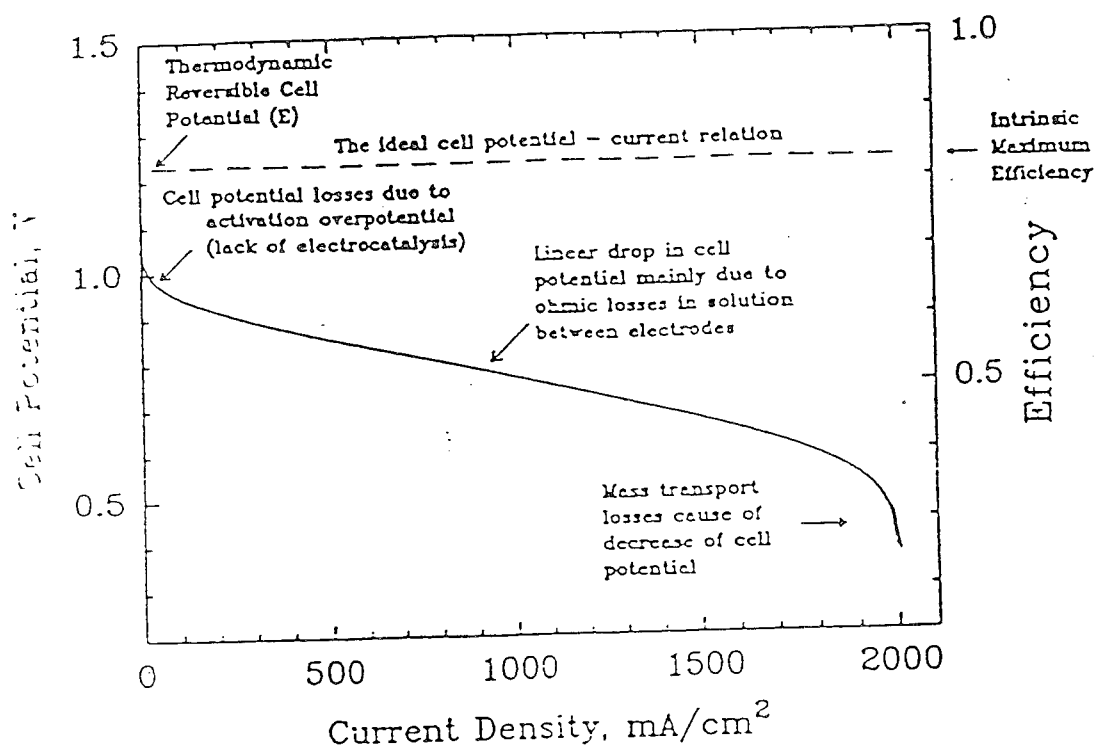
$$\epsilon_t = -\frac{nFE}{\Delta H} \quad (12)$$

$$\epsilon_r = -\frac{nFE}{\Delta G} \quad (13)$$

$$P = Ei \quad (14)$$

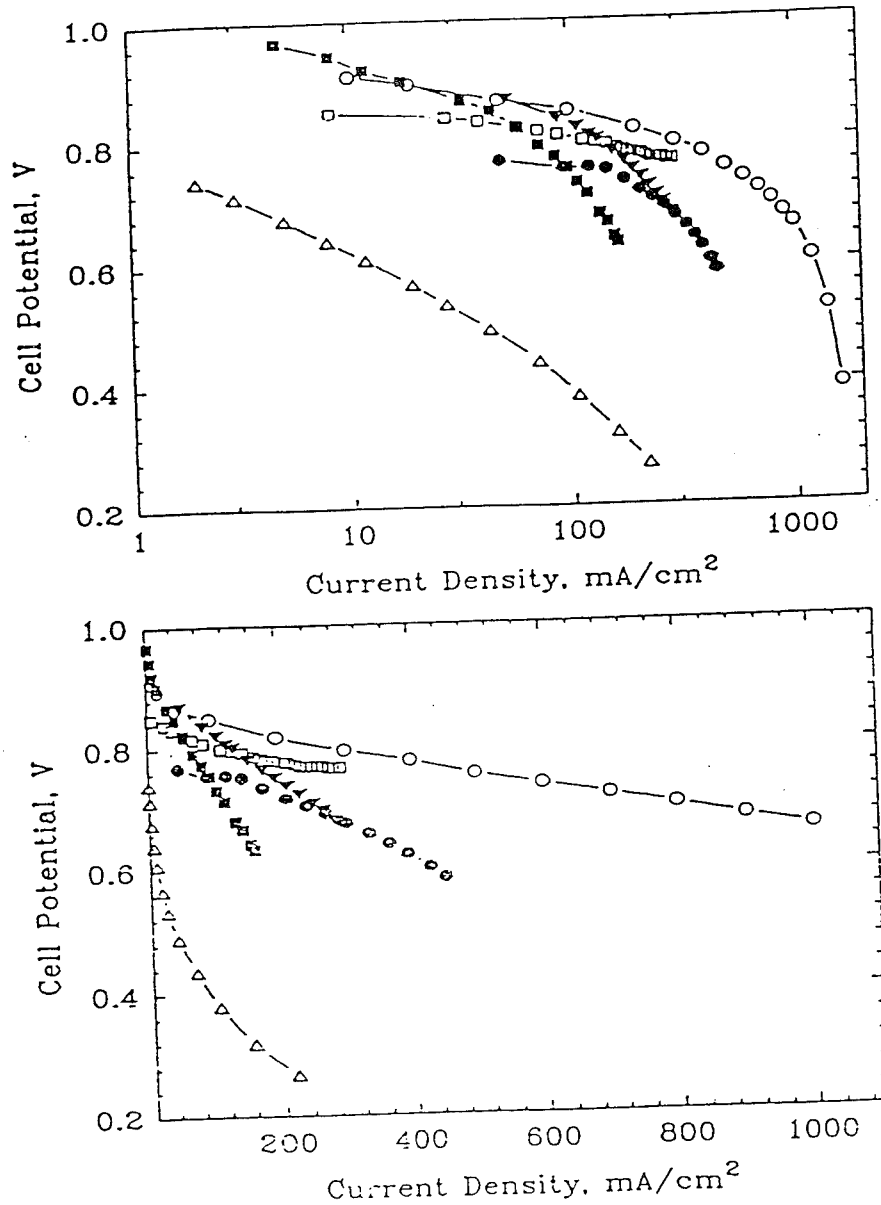
$$Q = -\frac{4.18 T \Delta Si}{nF} + i \sum \eta + i^2 R \quad (15)$$

Proton Exchange Membrane Fuel Cells From Basic Research to Technology Development

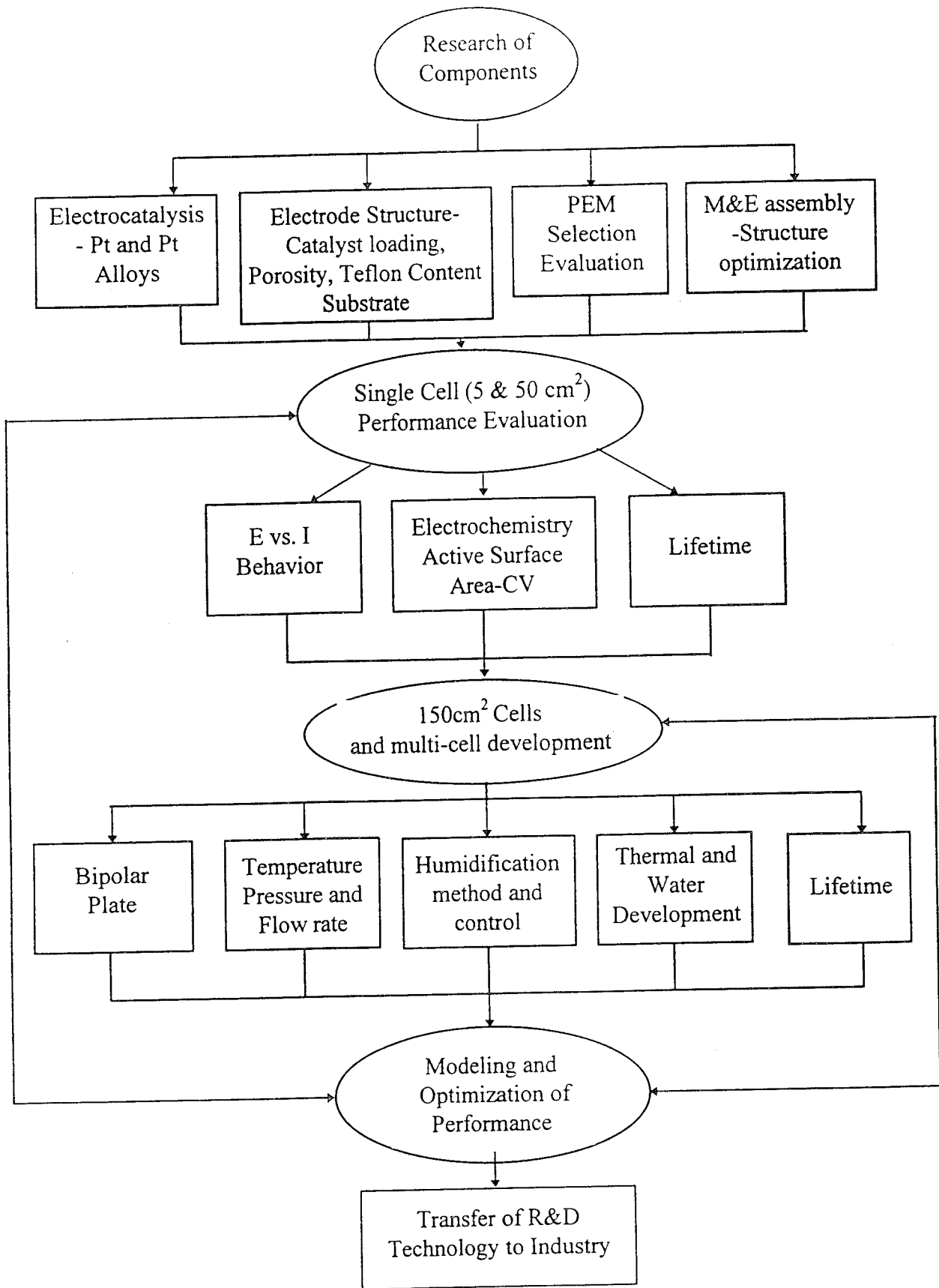


Typical potential vs. current density plot for a fuel cell illustrating the predominantly activation controlled, ohmic controlled and mass transport controlled regions.

Proton Exchange Membrane Fuel Cells From Basic Research to Technology Development

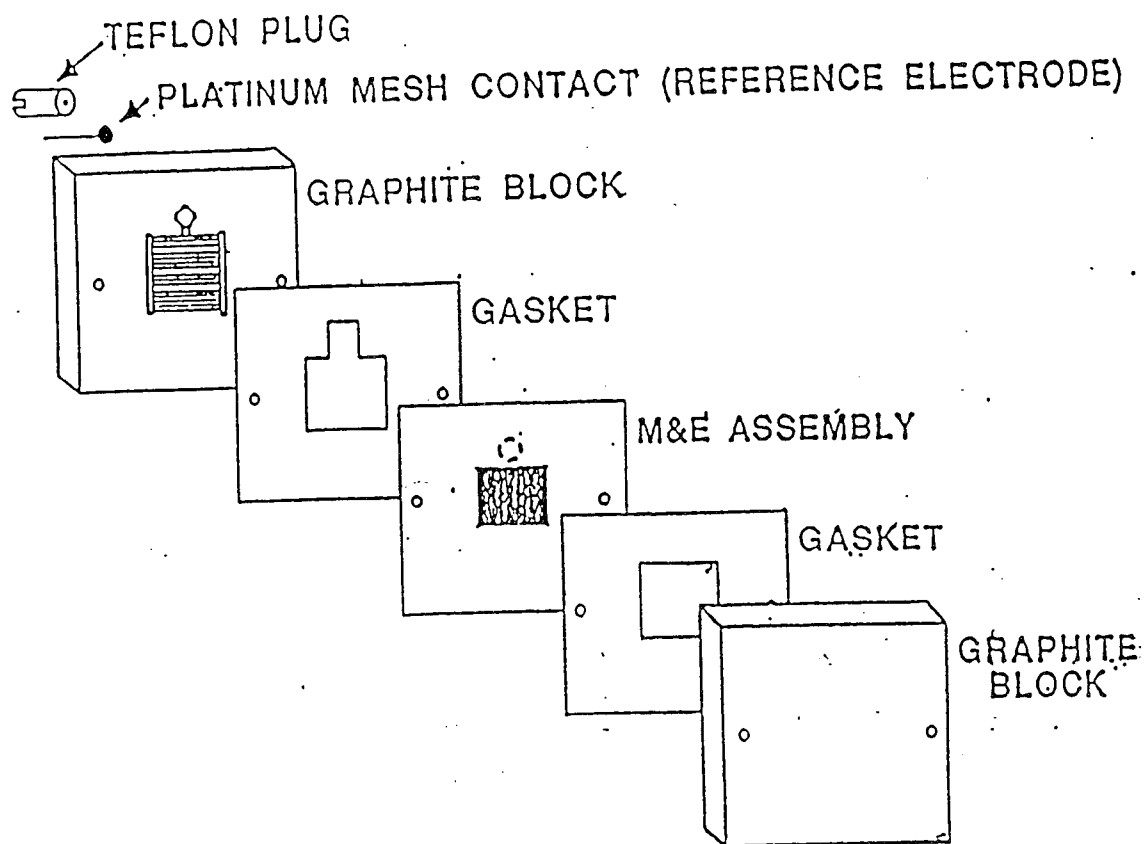


Potential vs. current density plots for state-of-the-art fuel cells:
 (○) PEMFC (CESHR); (●) SOFC (Westinghouse); (▼) MCFC (ERC);
 (□) PAFC - pressurized (Westinghouse); (■) AFC (BNL);
 (△) DMFC Calculated performance using best half cell data.



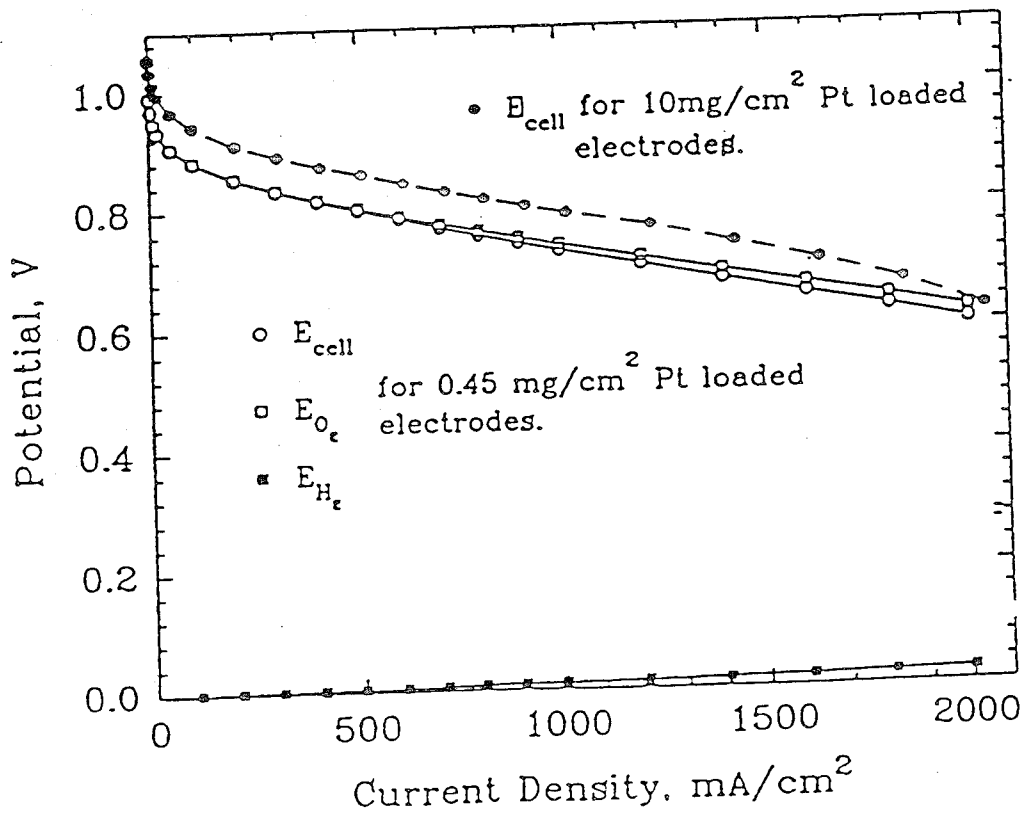
Flow-chart of proposed activities on the PEMFC project

Proton Exchange Membrane Fuel Cells *From Basic Research to Technology Development*

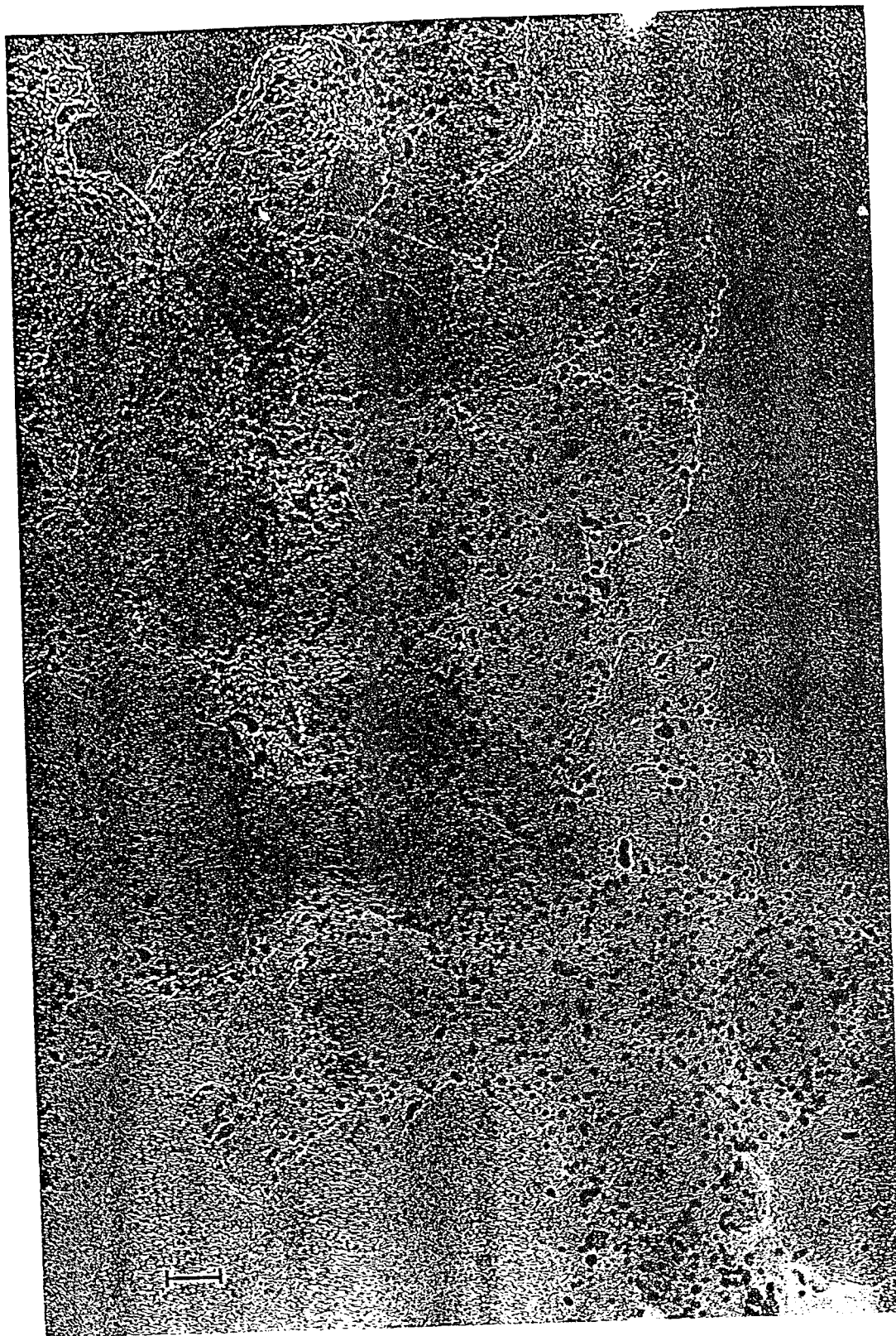


COMPONENTS OF SINGLE CELL 5 cm² ELECTRODES FOR SPE
HALF AND SINGLE CELL PERFORMANCE EVALUATION WITH
EXTERNAL HUMIDIFICATION OF REACTANTS

Proton Exchange Membrane Fuel Cells *From Basic Research to Technology Development*



Half cell and single cell performance in PEM H₂/O₂ fuel cell.



Microscopic photograph of carbon-supported Pt catalyst. (Scale bar = 2×10^{-6} cm.)

Low Pt-loading - High Performance

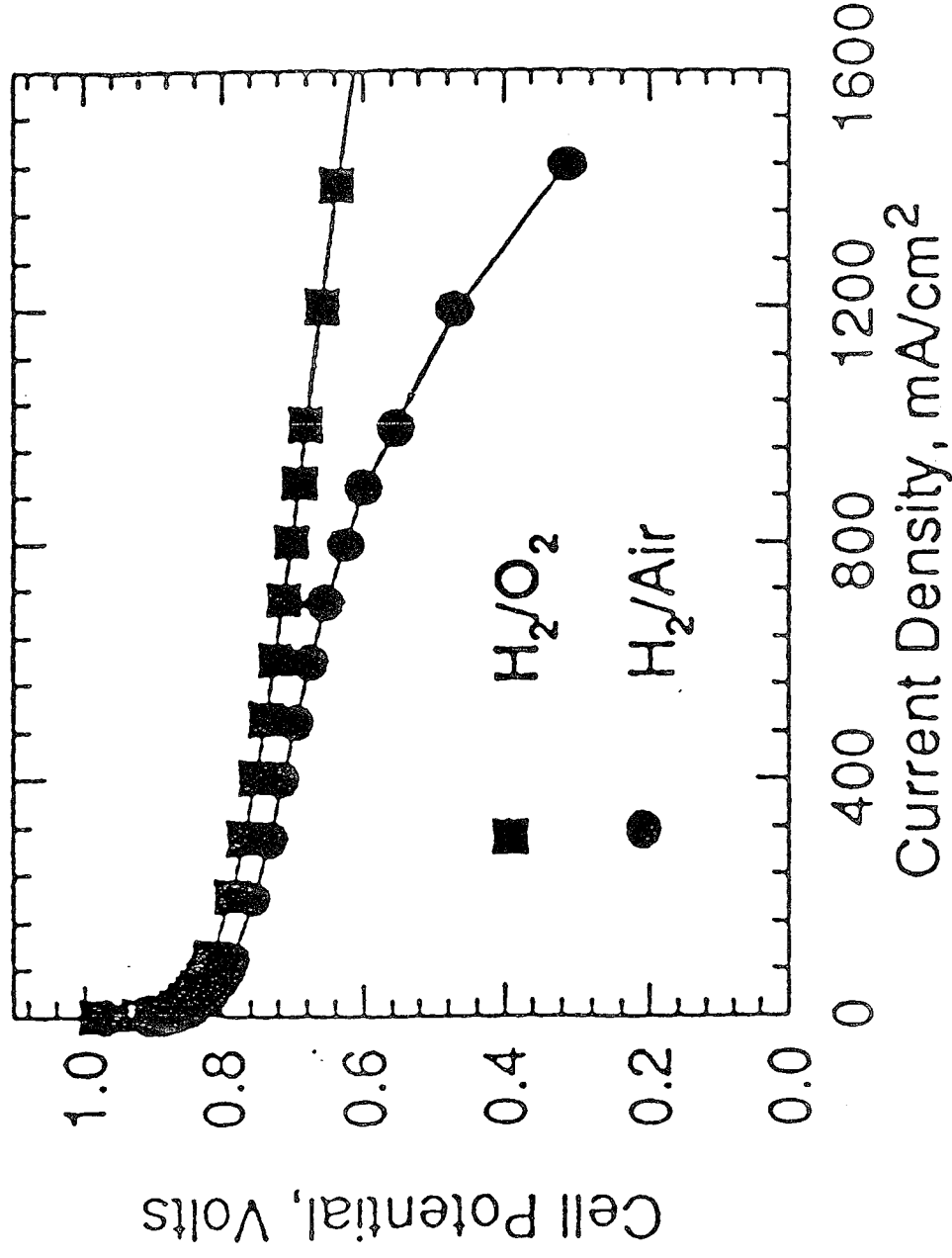


Figure 1 : Current Voltage Curves of H₂/O₂ and H₂/Air Single Cell. Operation at 1 bar, 70 °C. Dow membrane.

• Pt-loading 0.05 mg/cm².

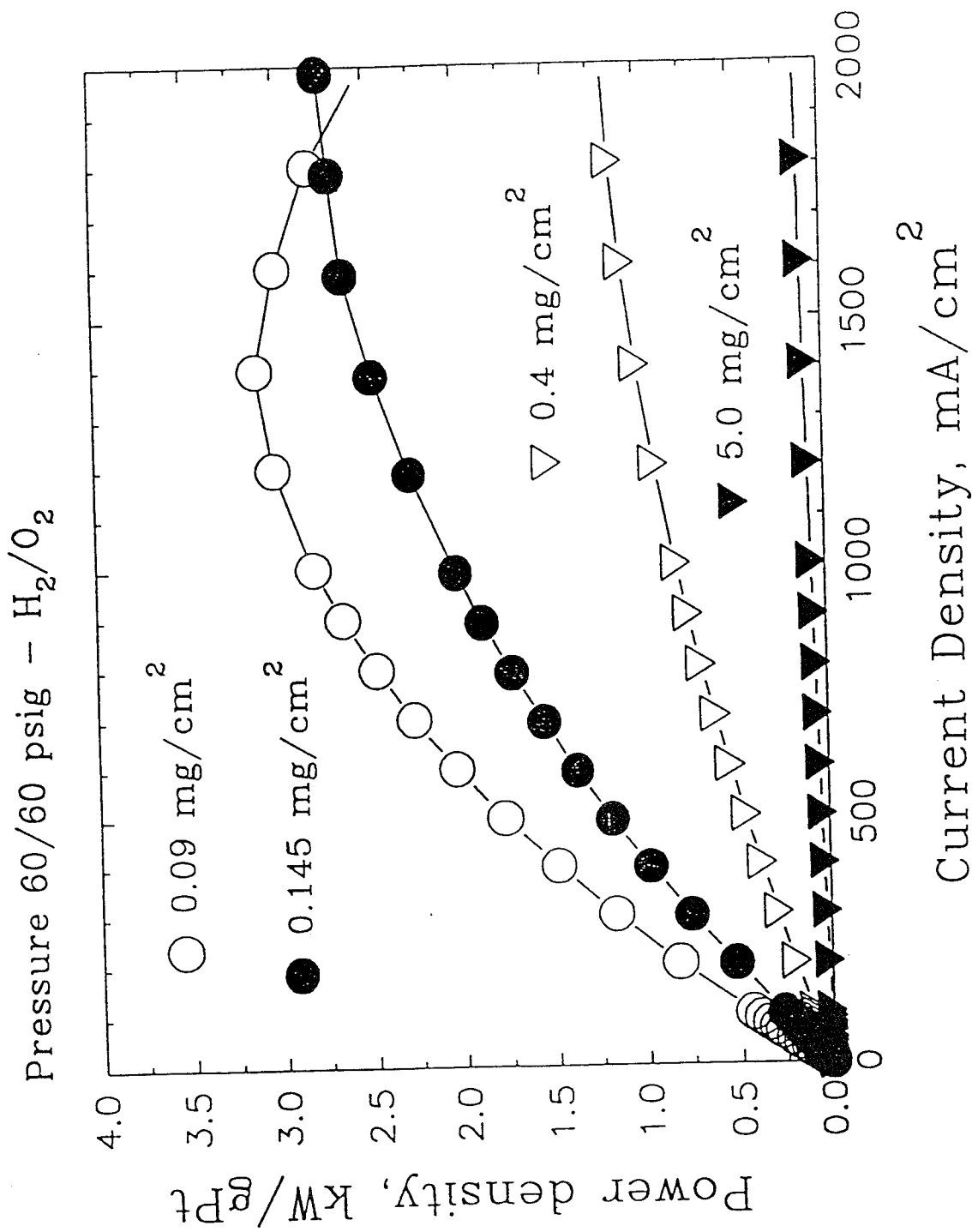
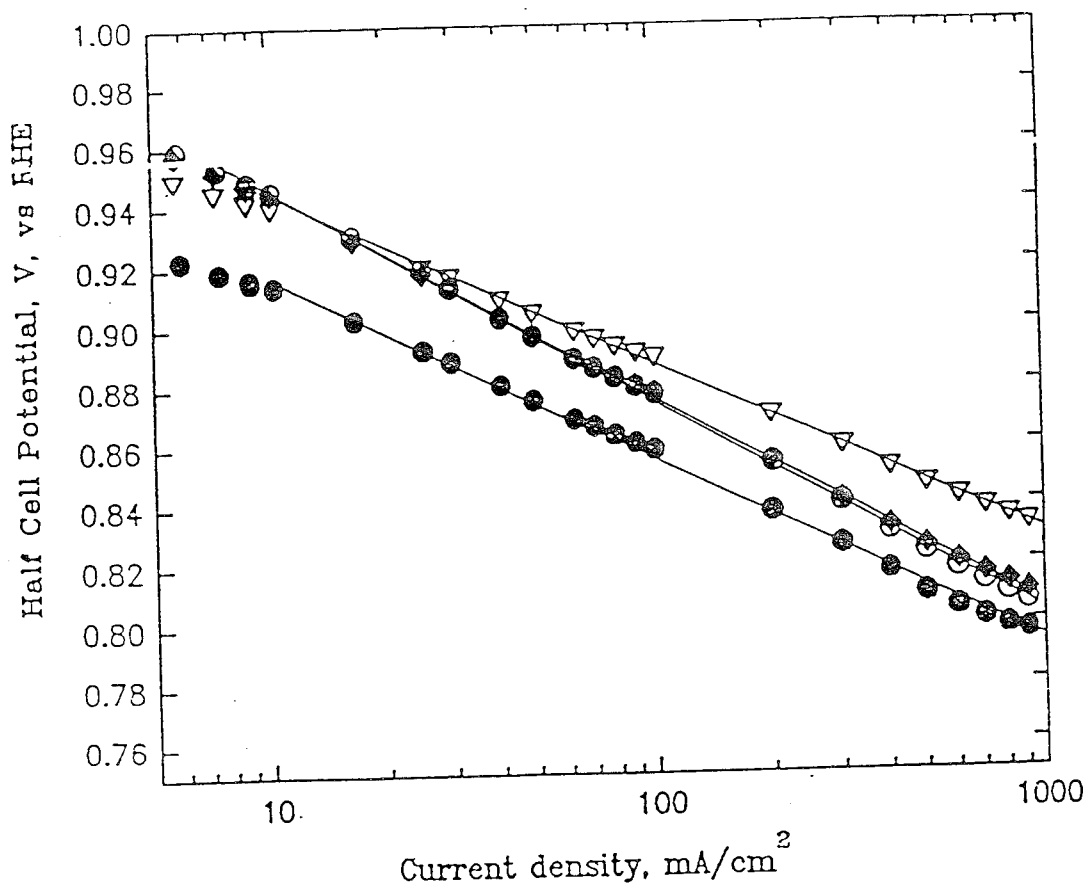


Figure 2 : Power delivered per g of Pt in each Electrode.
 (○) 0.09 mg/cm² CESHRR; (●) 0.145 mg/cm² CESHRR;
 (▽) 0.4 mg/cm² Etek Inc.; (▼) 5 mg/cm² Johnson & Matthey.

Proton Exchange Membrane Fuel Cells *From Basic Research to Technology Development*



iR corrected Tafel plots for oxygen reduction in proton exchange membrane fuel cells at 95°C and 5 atm on Pt and Pt alloy electrocatalysts, Pt loading in electrodes 0.3 mg/cm². Pt (●); Pt/Ni alloy (○); Pt/Co alloy (◆) and Pt/Cr alloy (▽).

Effect of Method of Preparation of Electrodes on Performance

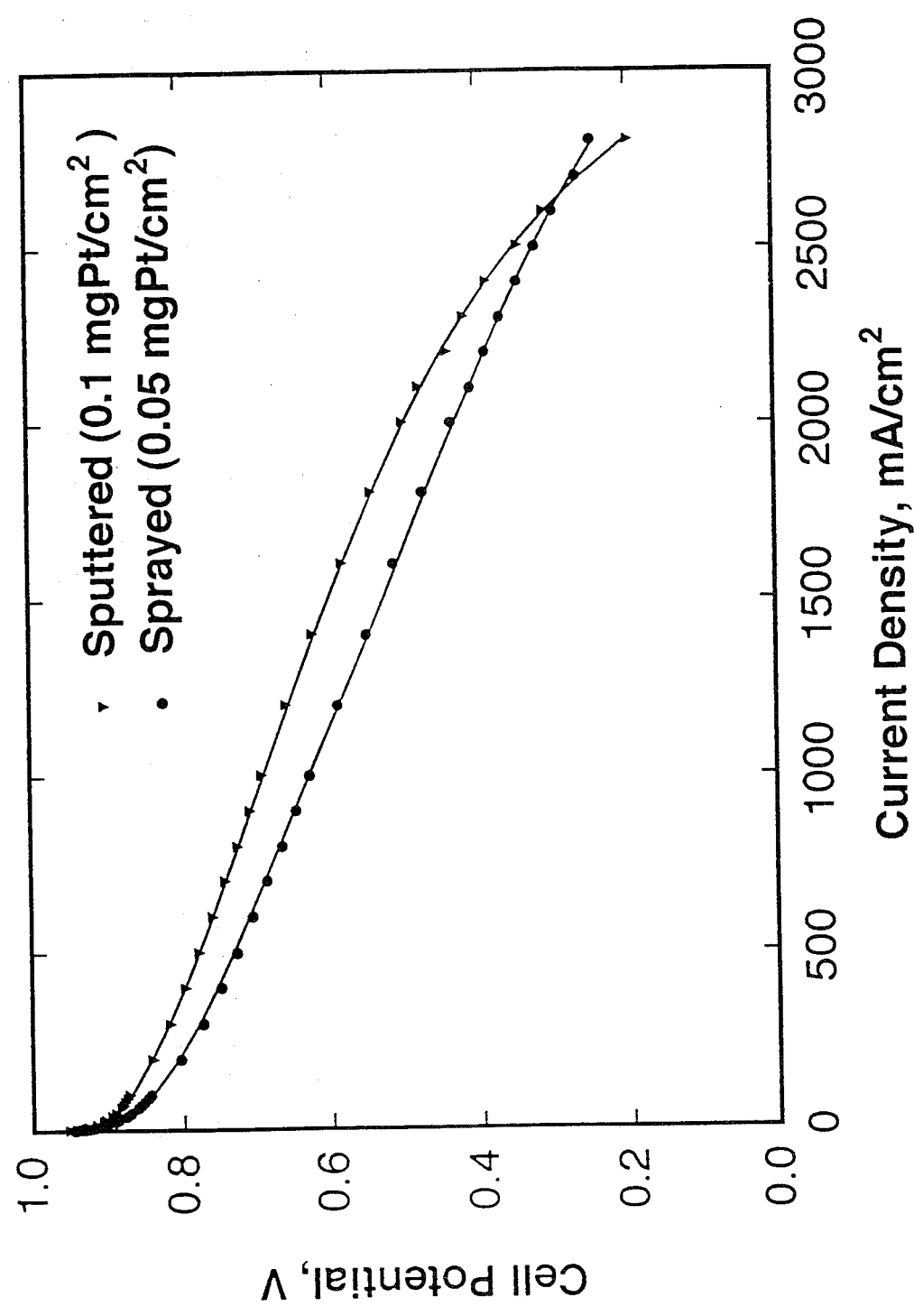
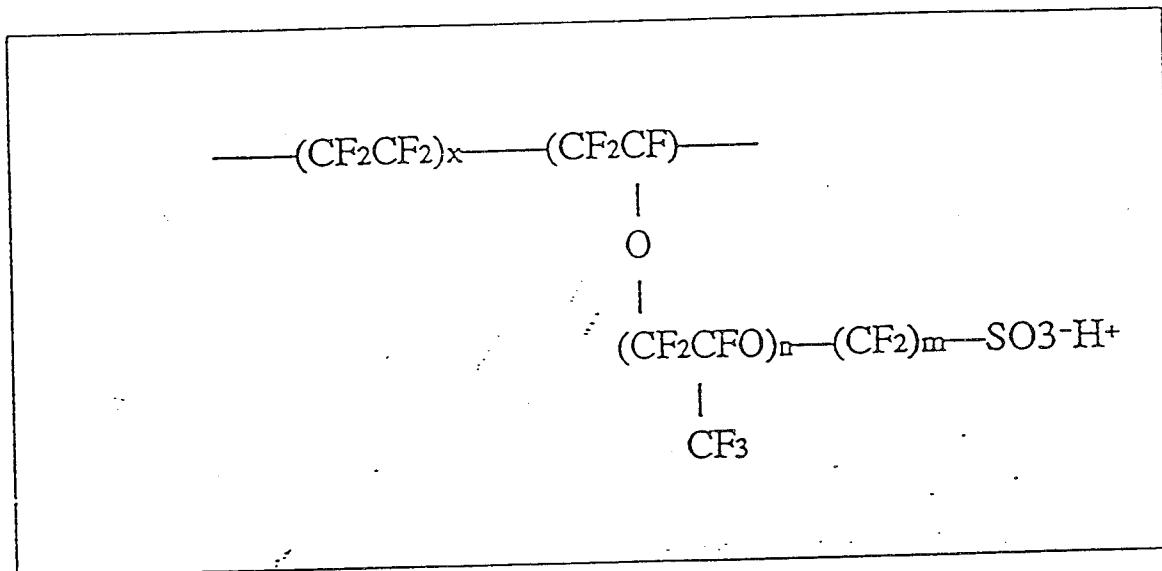


Figure 4 : Current-Voltage Curves of Single Cells with Cathodes prepared by the Spraying and Sputtering Techniques; H_2/O_2 , 5 bar, 95 °C

Proton Exchange Membrane Fuel Cells

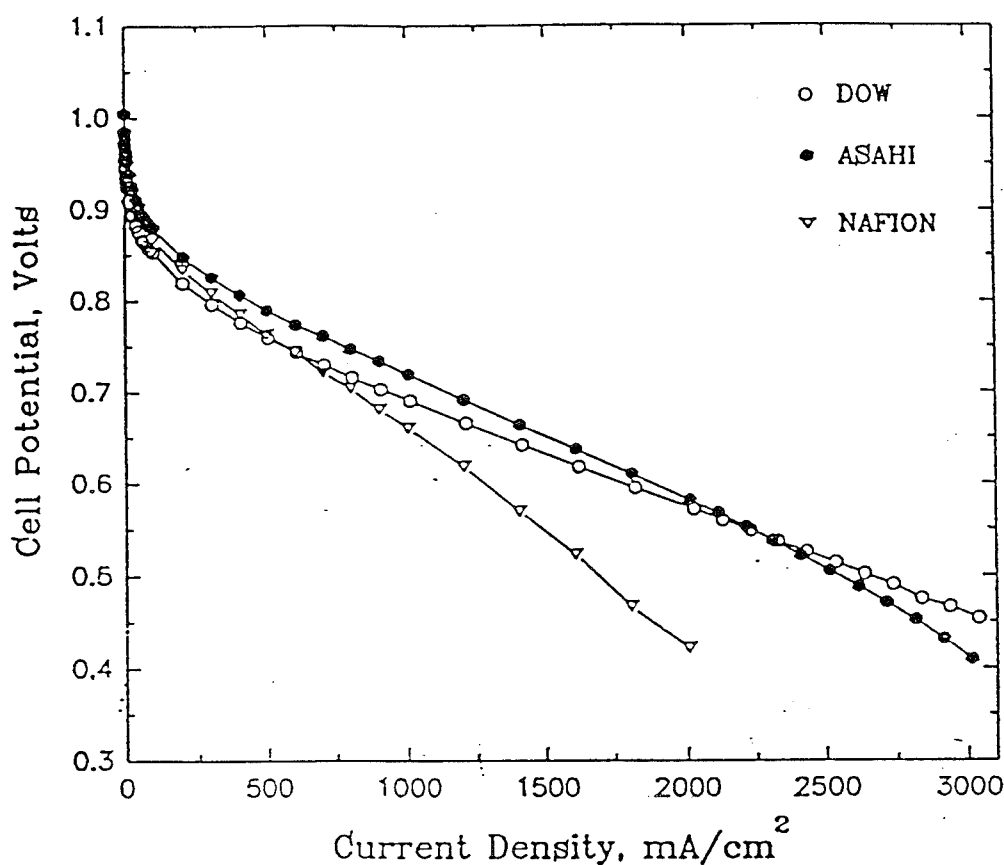
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Chemical Structure of Perfluorosulphonic Acid Polymer

	n	m	EW
Asiplex-S	0 ~ 2	2 ~ 5	1000
Dow	0	2	800
Nafion	1	2	1100

Proton Exchange Membrane Fuel Cells *From Basic Research to Technology Development*



Comparison of the cell performance with Asahi Aciplex[®]-S (120 μ m), Dow (125 μ m) and Nafion[®] (100 μ m) membranes. E-Tek electrodes (0.4 mgPt/cm²), pressure 5 atm, temperature 95°C, Hydrogen/Oxygen.

Analysis of Mass-Transport Effects

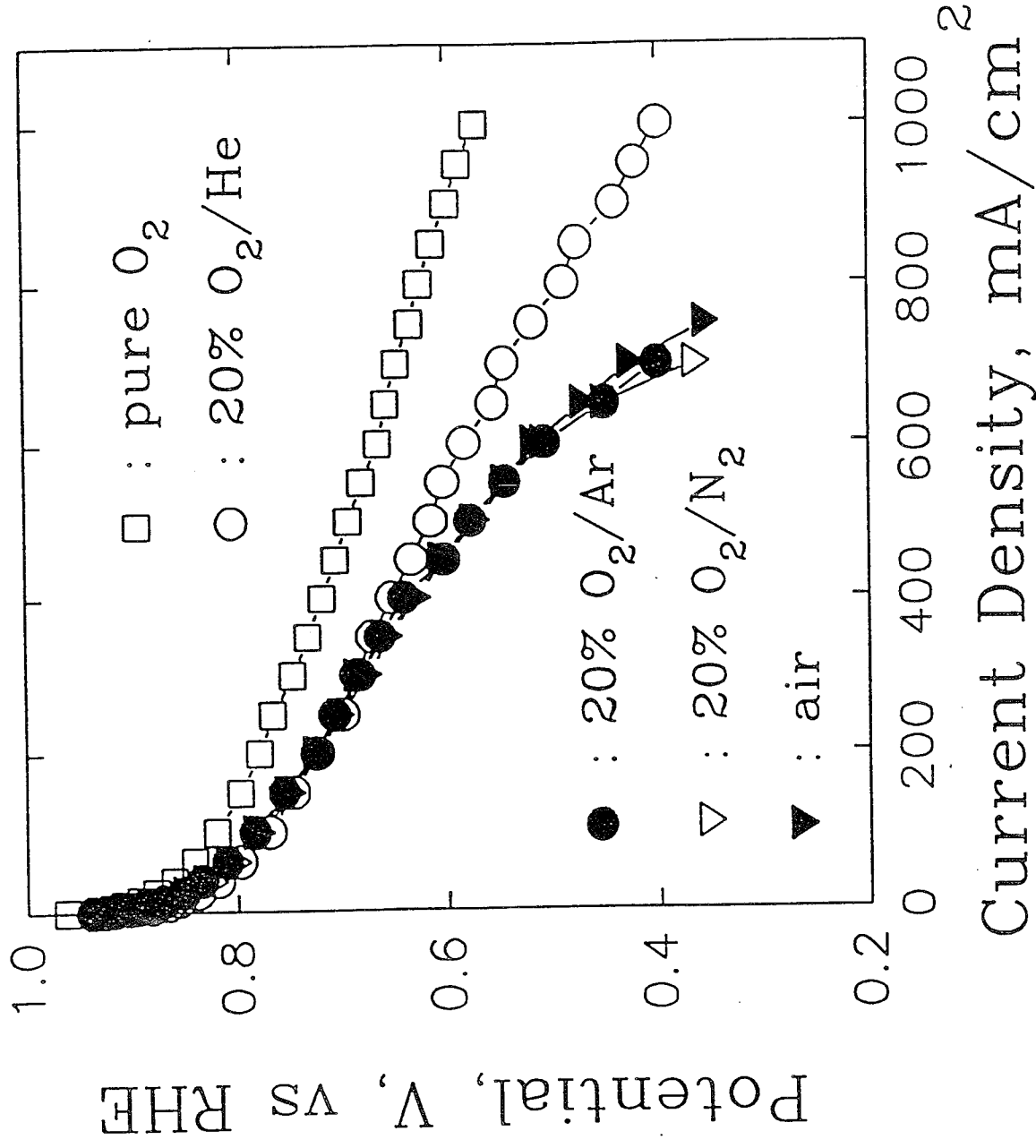
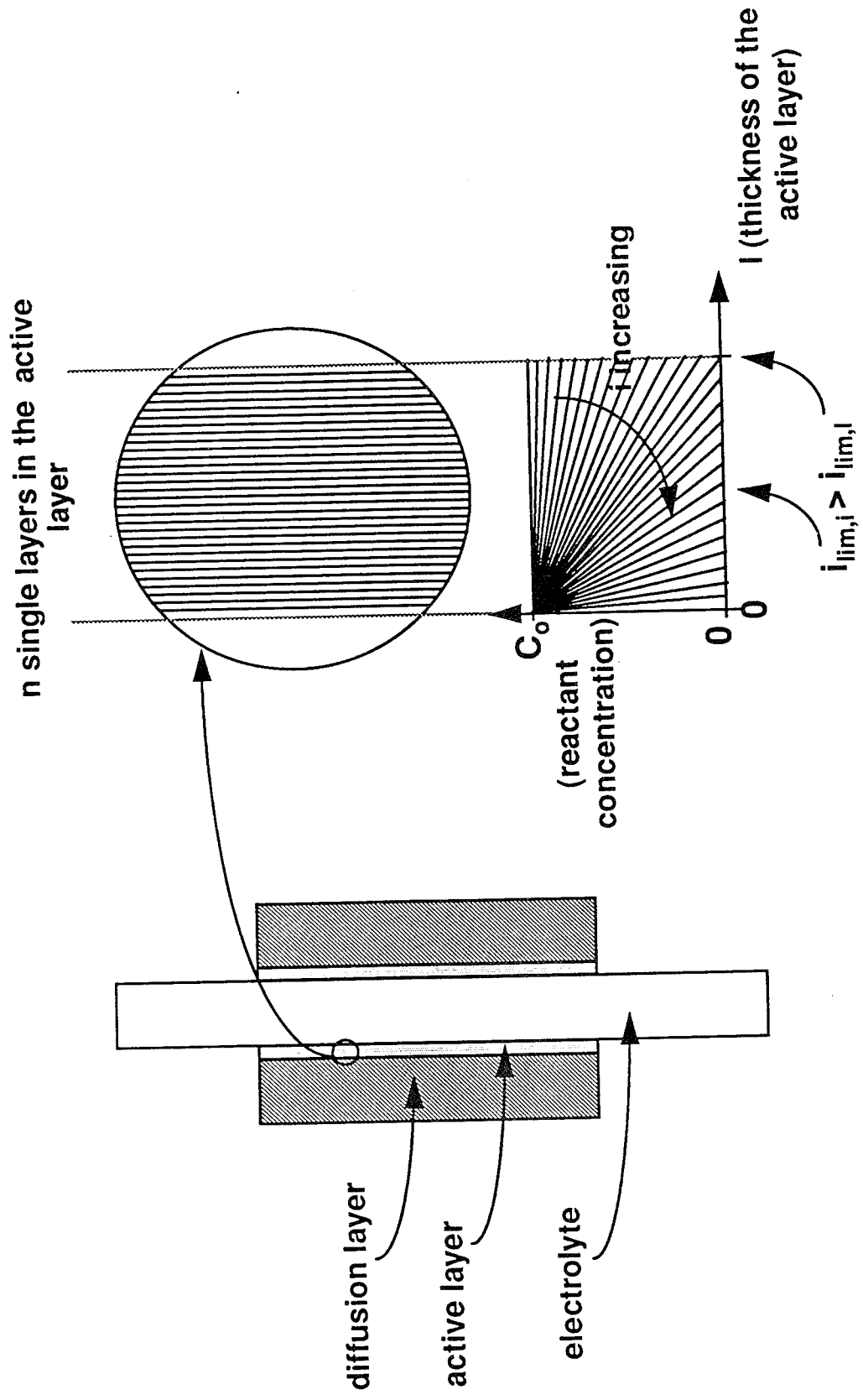
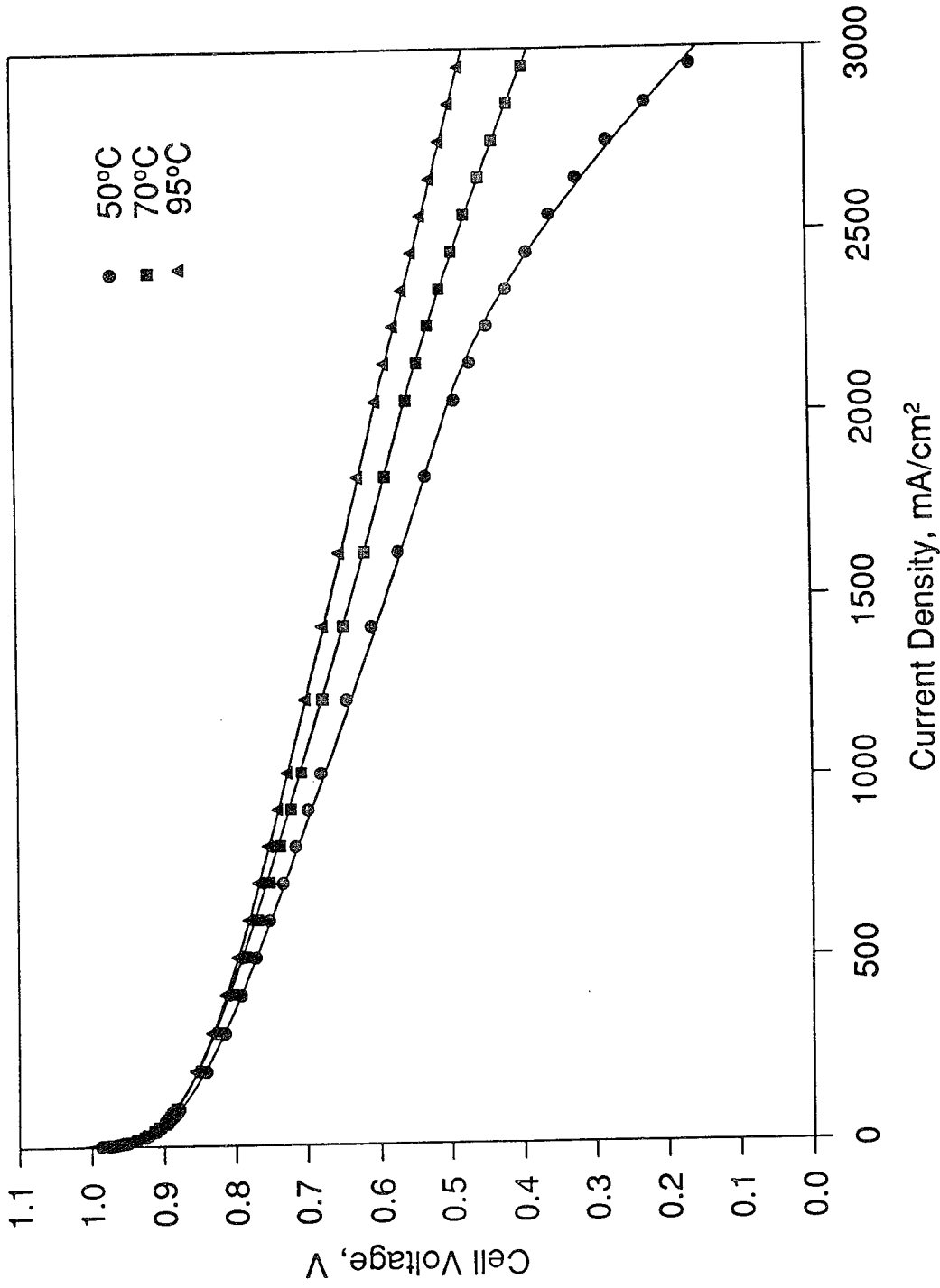


Figure 3 : Effect of different gas composition on the performance.
Operation at 70 °C and 1 bar.



Schematic of PEMFC Single Cell
and of Model for Finite Element Analysis of Mass Transport
in Active Layer of Electrode



**Modeling Study (lines) and Experimental Data (symbols)
for a H₂/O₂ PEMFC Running at 5/5 atm**

Operation *without* ext. Humidification

BCS Technology, Inc. Bryan, Texas

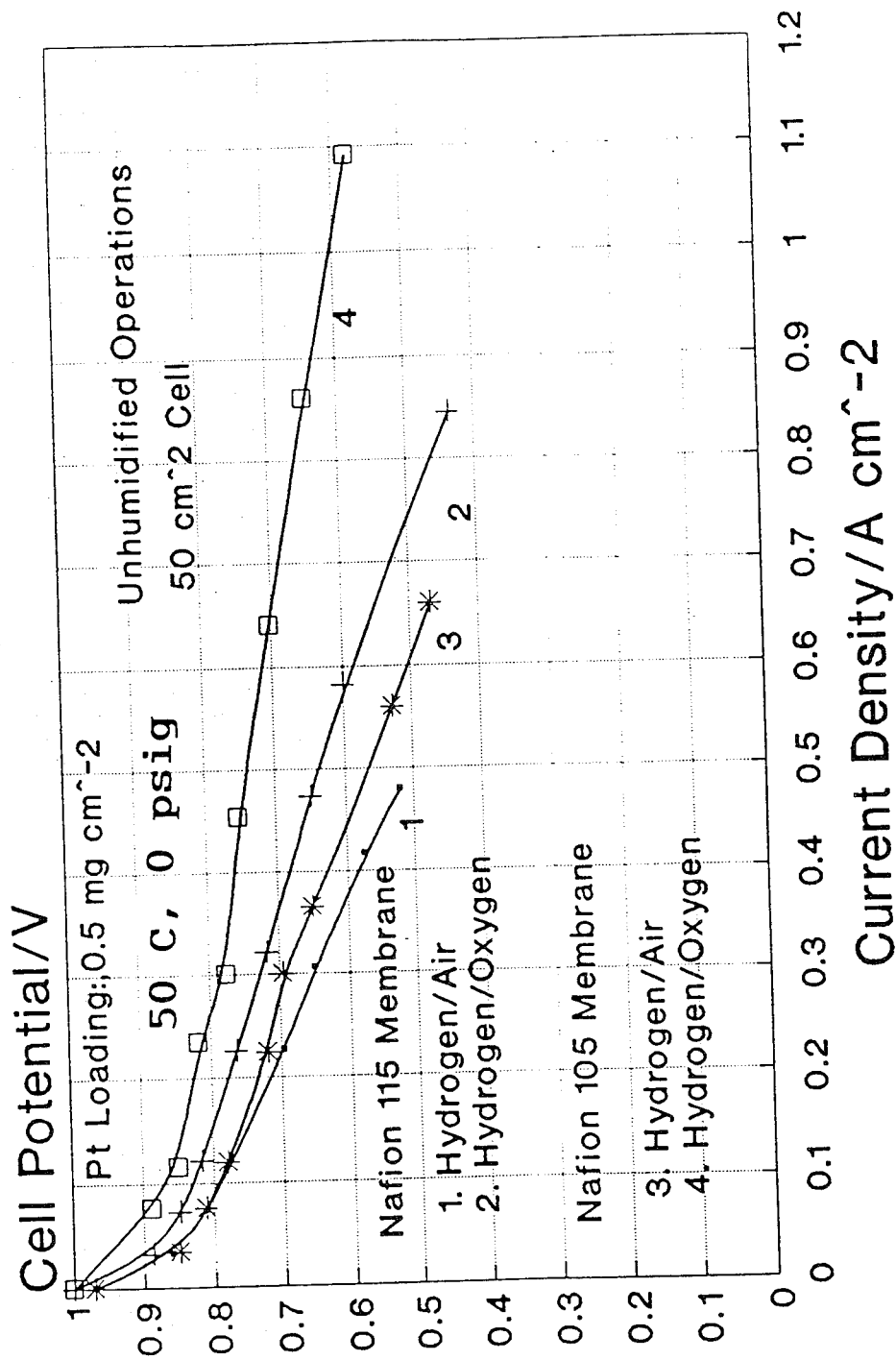


Figure 5 : Current Voltage Curves of 50 cm² H₂/O₂ and H₂/Air single cell, at 50 °C and gas pressure of 1 bar.

Low Pt-loading 4-Cell Stack

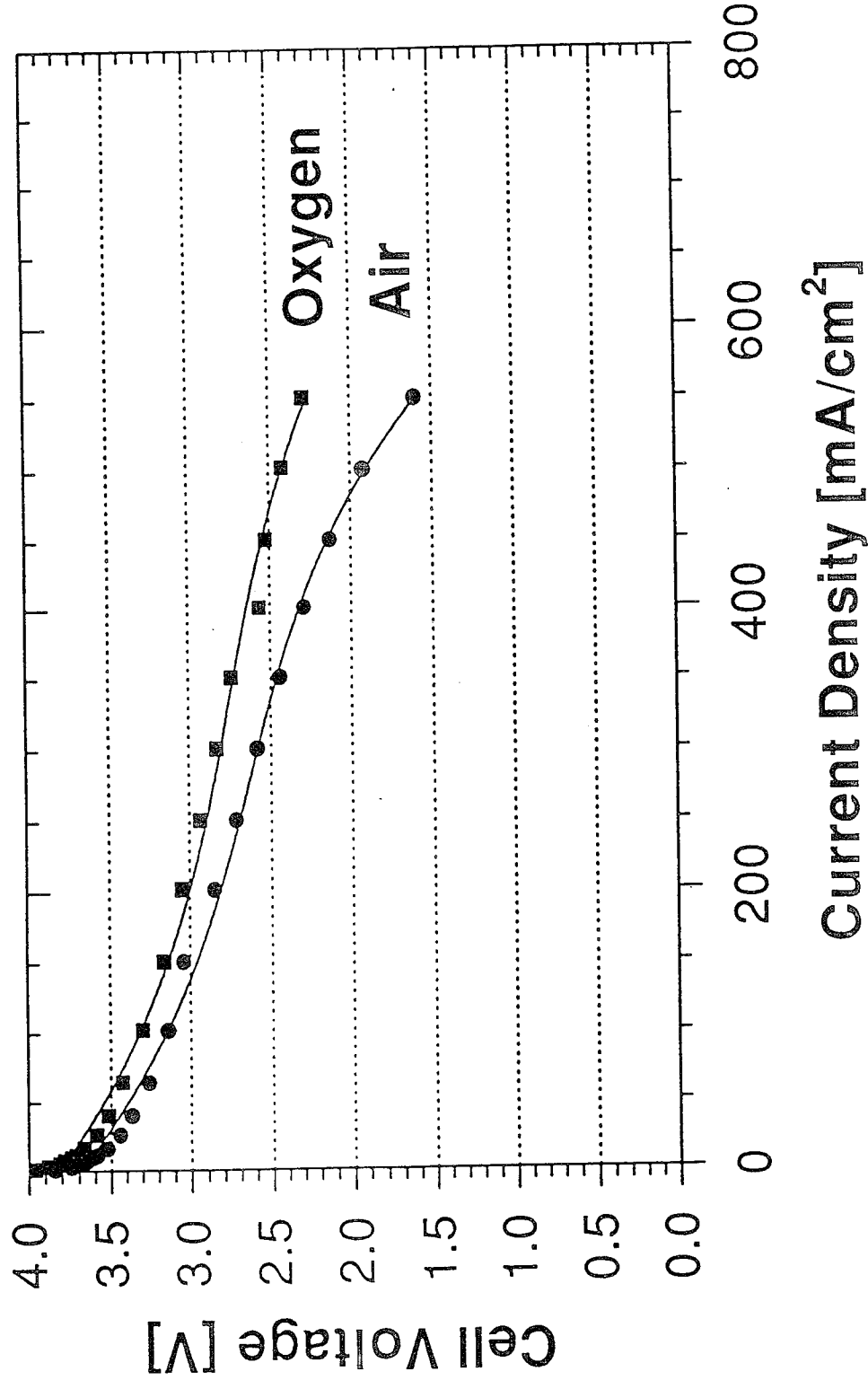
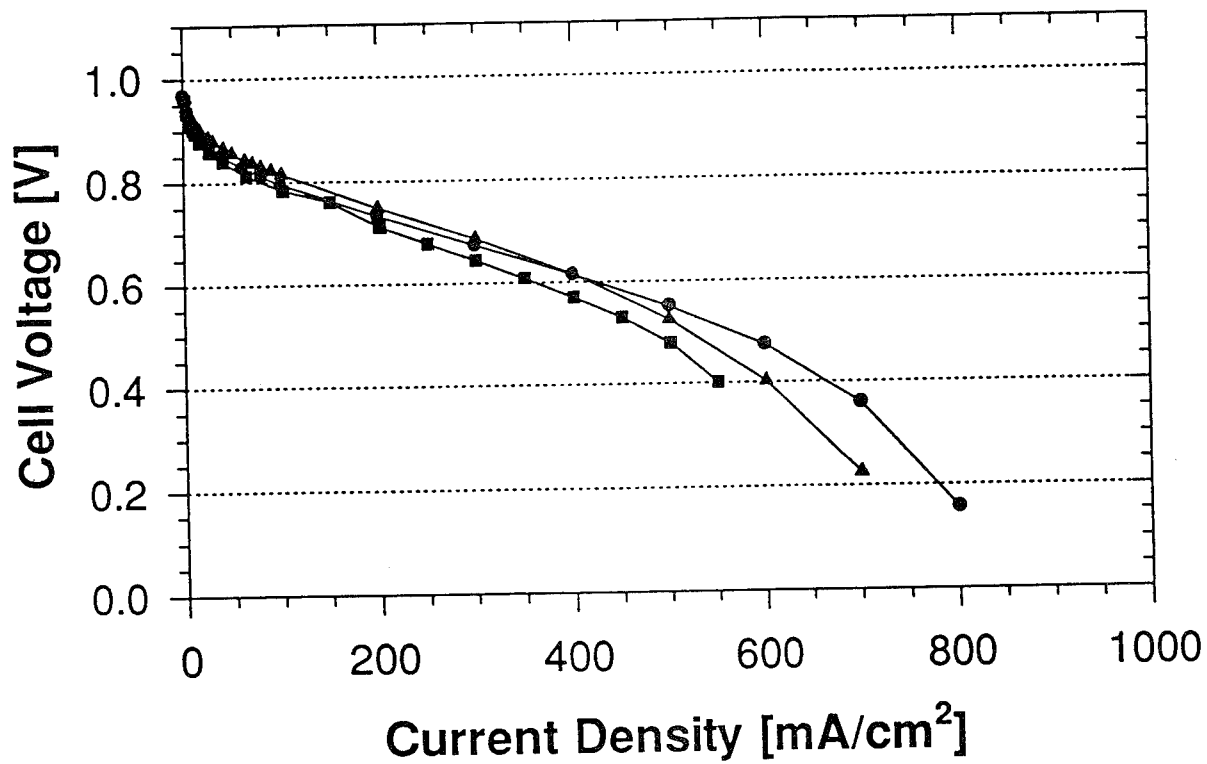


Figure 6 : Current Voltage Curves of 4 x 50 cm² Cell Stack. Operation at 3 bar, 50 °C. Pt-loading 0.4 mg/cm².



Current-Voltage Characteristic of 5 cm² and 50 cm² Single Cells and 50 cm² 4-Cell Stack. Hydrogen-Air at 3 bar, 50 °C.

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

CONCLUSIONS

- PEMFC - Most Promising Fuel Cell to Attain High Energy Efficiencies, High Power Densities and Long-life Times
- Pt Requirement Reducible to 0.2 g/kW
- Low-cost Membranes (\$50/m²) with Specific Conductance $> 10^{-1}$ ohm⁻¹ cm⁻¹ Vitrally Needed
- Optimization of M&E Structure Essential to Minimize Mass Transport for Operation with H₂/Air at 1 atm
- Operation with Internal Humidification - a Great Advantage
- Demonstration of Scale-up and Multicell Stack with Low Pt-loading Electrodes

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

TECHNO-ECONOMIC CHALLENGES

- Significantly Decrease Capital Costs, Weight and Volume
- Advanced Methods for Fuel Storage, Fuel Processing
- Minimize Mass Transport Overpotentials in H₂/Air Fuel Cells for Operation at 1 atm
- Lower Fabrication Costs of Electrodes
- Light-weight, Low Cost Bipolar Plates
- Alternate Membranes - Low Cost, Higher Temperature Operation
- Novel Methods for Water and Thermal Management
- DMFCs - Vital Need for Enhancing Electrocatalytic Activity by Several Orders of Magnitude and Inhibiting Methanol Cross-Over

**PROTON EXCHANGE MEMBRANE FUEL CELLS -
BASIC RESEARCH TO TECHNOLOGY
DEVELOPMENT**

ACKNOWLEDGEMENTS

- Former Coworkers
 - At LANL: E.A. Ticianelli, C.R. Derouin, W. Paik,
I.D. Raistrick, S. Gottesfeld, J.G. Beery and
T.E. Springer
 - At TAMU: E.A. Ticianelli, M.A. Enayetullah,
D.J. Manko, O.A. Velev, H.P. Dhar, H. Koch, A. Nandi,
D.H. Swan, A. Parthasarathy, S. Mukerjee, B. Dave,
S. Somasundaran and N.K. Anand

**PROTON EXCHANGE MEMBRANE FUEL CELLS -
BASIC RESEARCH TO TECHNOLOGY
DEVELOPMENT**

ACKNOWLEDGEMENTS

- Sponsors
 - At LANL: U.S. Department of Energy, NASA Lewis Research Center
 - At TAMU: U.S. Defense Advanced Research Projects Agency, NASA Johnson Space Center, NASA Lewis Research Center, NASA Sponsored Center for Space Power at TAMU, Texas Higher Education Coordinating Board, Energy Research Corporation, Asahi Chemical Industry Company, Ltd., Mazda Motor Corporation and Korea Gas Corporation

FUEL CELL DEVELOPMENT PROGRAM
for
U.S. ARMY

PROSPECTOR VII

OCTOBER 1994

ANALYTIC POWER CORPORATION
PO BOX 1189
BOSTON, MA 02117

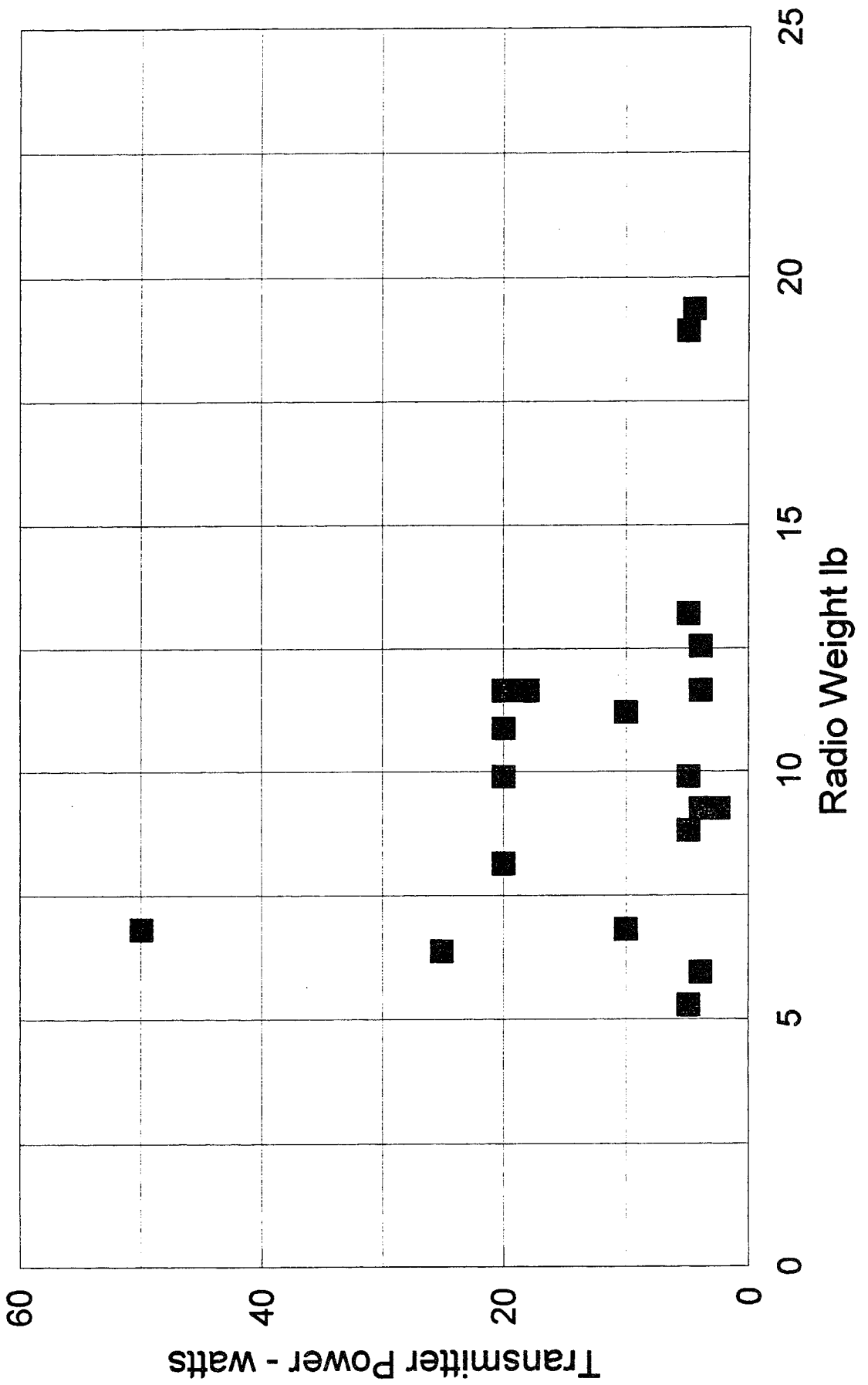
(617) 542-6352
(617) 695-3272 FAX

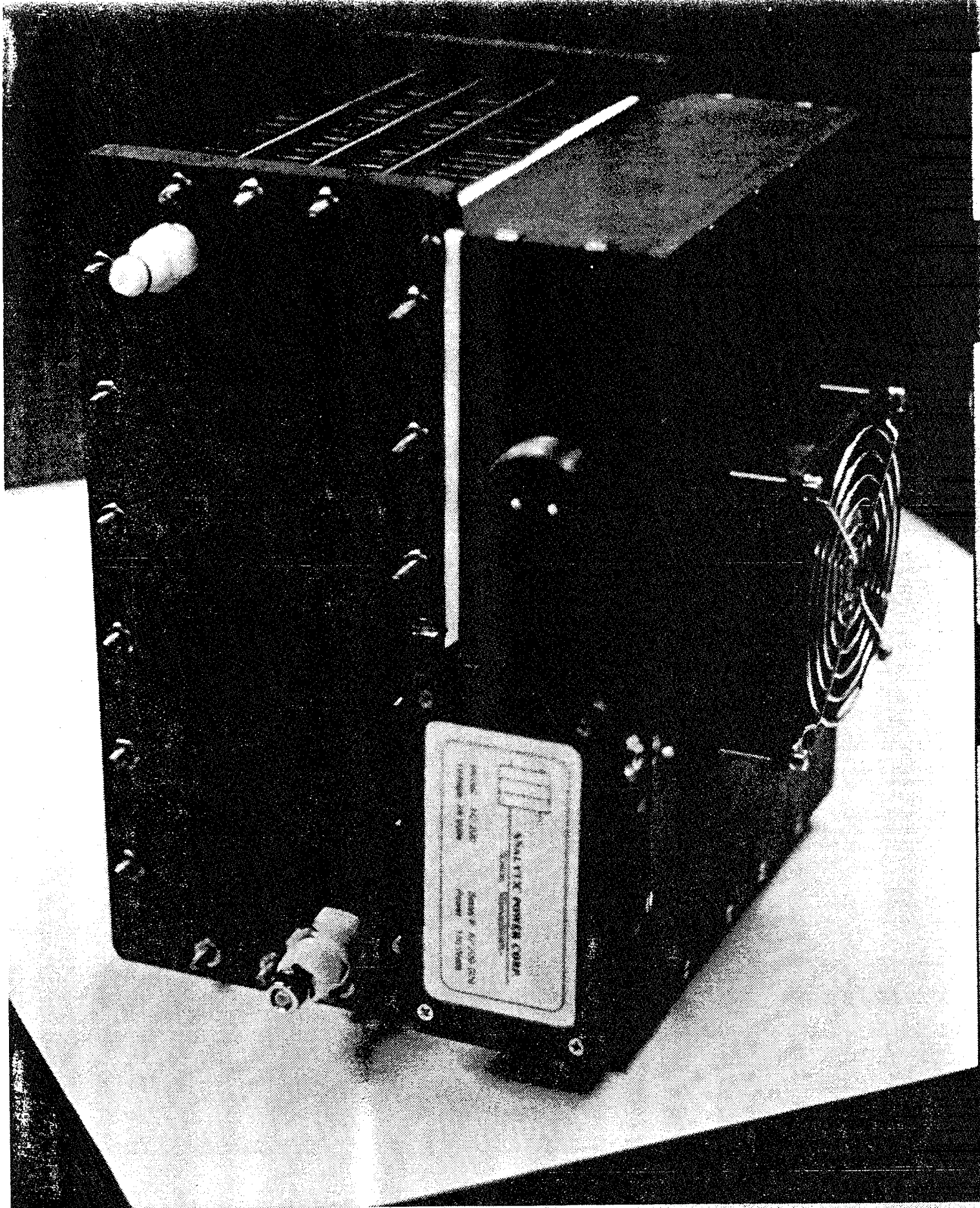
U.S.Army Batteries APPLICATIONS

TRANSMITTER	Raytheon, GD, ITPA & LASER	Hughes, TRW		
AMP	4	5	3	6
VOLT	24	24	12	24
WATTS	96	120	36	144
SOLDIER SYSTE(21 CLW)	RANGE FINDERS (700-800 SHOT			
AMP	12.5	AMP 0.5		
VOLT	24	VOLT 24		
WATTS	300	WATTS 12		
Radio Beacon	DESIGNATORS (50-100 SHOTS)			
AMP	33	17.5	6	15
VOLT	12	24	48	24
WATTS	396	420	288	360

Transmitter Power vs Weight

Army Man Pack Radios





ANALYTIC POWER CORPORATION
 TYPE 4.2.0 FUEL CELL STACK
 Forced Convection/ambient
 SMALL FORMAT

POWER 153 watts
 VOLTAGE 28 volts
 CURRENT 5 amp
 CELL VOLTAGE 0.723 volts
 CURRENT DENSITY 125 ASF
 No. OF CELLS 39
 CELLS/COOLER 1
 CAT LOAD. 0.50 mg/cm²

	W(in)	L(in)	AREA(in ²)
ACTIVE	2.50	2.50	6
TOTAL	4.00	3.75	15

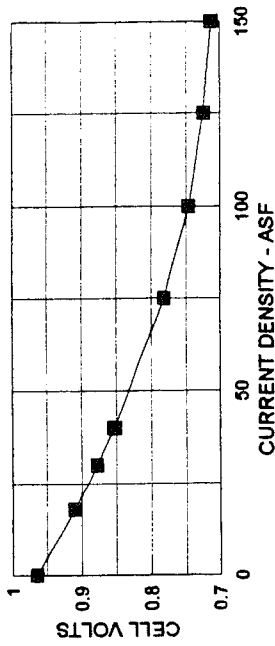
ANALYTIC POWER CM-4a CELL
 DATE June 28, 1984

POWER DENSITY 1.00 watts/in³
 18.38 watts/lb
 LENGTH in 10.2 in
 WEIGHT lb 8.3 lb
 VOLUME 153 in³

MATL. COST (\$) \$4,265
 MATL. COST (\$/watt) \$28
 TOT. COST (\$) \$9,420
 TOT. COST (\$/watt) \$62

STACK 4.2 PERFORMANCE

ANALYTIC POWER CORP.

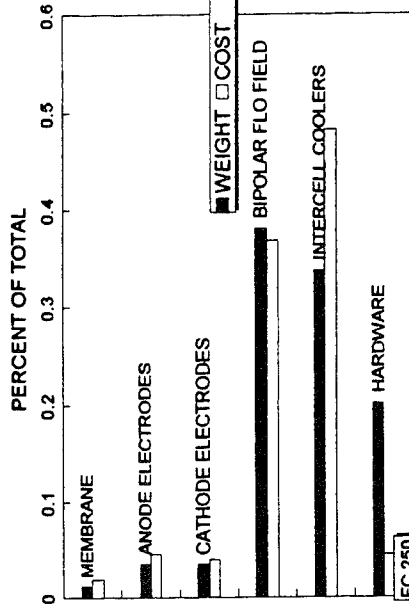


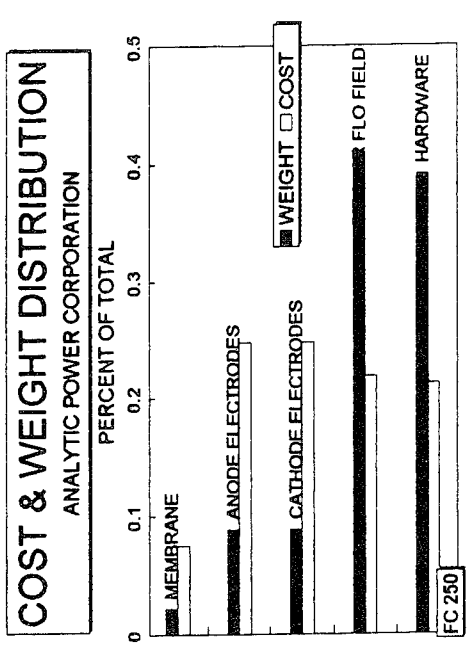
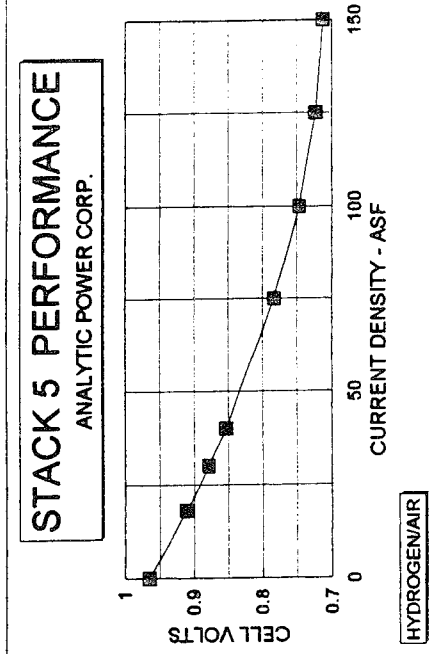
HYDROGEN/AIR

COST & WEIGHT DISTRIBUTION

ANALYTIC POWER CORPORATION

CELL COMPONENTS	MATERIAL	WEIGHT (lb)	WEIGHT (%)	COST (\$)	COST (%)
MEMBRANE		0.10	1.2%	\$80	1.9%
ANODE ELECTRODES		0.29	3.5%	\$195	4.6%
CATHODE ELECTRODES		0.29	3.5%	\$169	4.0%
BIPOLAR FLO FIELD		3.17	38.1%	\$1,570	36.8%
INTERCELL COOLERS		2.81	33.7%	2058.88	48.3%
HARDWARE		1.67	20.1%	\$192	4.5%
		8.326	100.0%	\$4,265	100.0%





ANALYTIC POWER CORPORATION		ANALYTIC POWER CM-4a CELL	
TYPE 5 FUEL CELL STACK		DATE Aug 26, 1994	
Forced Convection/ambient/No Carbon mini FORMAT			
POWER	153 watts	POWER DENSITY	3.25 watts/in ³
VOLTAGE	24 volts		61.36 watts/lb
CURRENT	7 amp	LENGTH in	5.2 in
CELL VOLTAGE	0.713 volts	WEIGHT lb	2.5 lb
CURRENT DENSITY	150 ASF	VOLUME	47 in ³
No. OF CELLS	33		
CAT LOAD.	0.50 mg/cm ²	MATL. COST (\$)	\$242
		MATL. COST (\$/watt)	\$2
ACTIVE	W(in) L(in) AREA(in ²)	TOT. COST (\$)	\$559
	2.50 2.50 6	TOT. COST (\$/watt)	\$4
TOTAL	3.00 3.00 9		

CELL COMPONENTS	MATERIAL	WEIGHT (lb)	WEIGHT (%)	COST (%)	COST (\$)
MEMBRANE		0.05	2.2%	7.5%	\$18
ANODE ELECTRODES		0.22	8.9%	24.7%	\$60
CATHODE ELECTRODES		0.22	8.9%	24.7%	\$60
BIPOLAR FLO FIELD		1.02	41.0%	21.8%	\$53
HARDWARE		0.98	39.1%	21.2%	\$51
		2.496	100.0%	100.0%	\$242

PROPRIETARY

BAR GENERATOR		2500 WHR	
ENDURANCE 16.33 HOURS			
composition	%wt	wt lb composition	%wt
NaBH ₄	55.0%	1.4	76.9%
P2O ₅	34.3%	0.9	5.8%
Binder	10.7%	0.3	17.3%
	100.0%	2.5	99.9%
DENSITY	0.8 in ³ /gmH ₂		
VOLUME	102.7 IN ³		

ANALYTIC POWER CORPORATION
 HYDROGEN & OXYGEN SOURCES
 MISSION 2.50E+03 watt hours

Weight - lb hydrogen	oxygen source	O2 GAS 14.9	CRYO O2 37.4	HNO3 9.5	KO2** 8.0	H2O2 5.5	NaClO3 6.8
H2 GAS	5.2	20.1	42.6	14.7	13.2	10.8	12.0
CRYO H2	3.6	18.5	41.1	13.1	11.7	9.2	10.4
MET HYDR	50.0	64.9	87.4	59.5	58.0	55.5	56.8
NaBH4	1.4	16.3	38.8	10.9	9.4	6.9	8.2
CaH2	3.1	17.9	40.5	12.6	11.1	8.6	9.9
BAR	2.7	17.6	40.1	12.2	10.7	8.3	9.5
Al/NaBH4	1.7	16.6	39.1	11.2	9.7	7.3	8.5
N2H4	2.3	17.2	39.8	11.8	10.4	7.9	9.1
NaBH4**	4.0	18.9	41.4	13.5	12.0	9.6	10.8
CaH2**	5.7	20.6	43.1	15.2	13.7	11.3	12.5
Al/NaBH4**	4.1	19.0	41.5	13.6	12.1	9.7	10.9

** water adc lb

Volume in3 hydrogen	oxygen source	O2 GAS 197	CRYO O2 531	HNO3 114	KO2** 328	H2O2 107	NaClO3 75
H2 GAS	382	578	912	495	710	488	456
CRYO H2	179	375	709	292	507	285	253
MET HYDR	156	353	687	270	484	263	231
NaBH4	39	235	570	153	367	145	113
CaH2	45	241	576	159	373	151	119
BAR	56	252	586	169	384	162	130
Al/NaBH4	26	222	556	139	353	132	100
N2H4	65	262	596	179	393	172	140
NaBH4**	112	309	643	226	440	219	187
CaH2**	118	315	649	232	446	225	193
Al/NaBH4**	92	289	623	206	420	199	167

** water adc in3

**Army Mission 2.5 kwh
Max Power 150 watts**

Specific Properties

Analytic Power

Power Source	\$/kWH	whr/lb	whr/in3
Honda EM 350	\$178	101	1.9
BA5590	\$376	73	3.0
BA5699	\$3,516	72	2.7
Li IO _n	?	114	3.0
GelCel	\$66	55	4.5
NiCad	\$2,105	15	1.6
FC-200 & Fuel Pac	\$3,256	186	6.1
STK 5x Fuel Pac	\$464	510	17.2

Mission Properties

Power Source	\$	lb	in3
Honda EM 350	\$445	24.7	1341
BA5590	\$940	34.5	833
BA5699	\$8,790	34.7	919
Li IO _n	?	21.9	833
GelCel	\$165	45.5	556
NiCad	\$5,263	168.2	1543
FC-200 & Fuel Pac	\$8,141	13.5	413
STK 5x Fuel Pac	\$1,159	4.9	145

OVERVIEW OF PEM FUEL CELL RESEARCH AT ARL

MICHAEL BINDER
ARL, POWER SOURCES DIVISION
FORT MONMOUTH, NJ 07703

NOVEMBER 1, 1994

OVERVIEW OF PEM FUEL CELL RESEARCH AT ARL

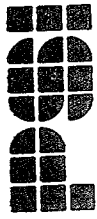
<300W Portable Fuel Cell

- Hydrogen Generation
 - Irreversible Alkali Hydride (PSI)
- Improved Electrocatalysts
 - Catalyst/Support
 - Modify Carbon Blacks
 - Pt and Pt-Ru Catalysts
- PEM Characterization/Evaluation
 - Water Uptake
 - Conductivity
 - Thermal Measurements
- Improved Solid Electrolytes
 - Modification
 - Electron Beam (Dupont)
 - Gas Plasma
 - Gamma Rays
 - Synthesis
 - Polyphenylenes (Maxdem)
 - Sulfonated PEEK (JPL)

HYDROGEN GENERATION

Irreversible Metal Hydride

- Physical Sciences, Andover, MA
- Deliver prototype portable hydrogen source by Jan 95
- >90% hydride utilization



PROTOTYPE PROGRAM GOALS

T-17334

- Overall goal
 - Design, test and deliver prototype portable hydrogen source

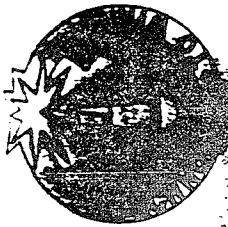
- Prototype performance characteristics
 - 3 Kwh energy capacity
 - 4 kg weight
 - 6.5ℓ volume
 - 50 to 250W

- Hydrogen generated by alkali hydride reaction with water
 - $\text{LiH} + \text{Al} + 2\text{H}_2\text{O} \rightarrow \text{LiAlO}_2 + 5/2 \text{H}_2$
 - High mass fraction hydrogen (7%)
 - Reduced reactant passivation

Catalyst / Support

Modify Carbon Black

- Carbon Black Catalyst Supports Contain Various Functionalities
- Use Gas Plasma Treatment to Modify Material Properties
 - Rapid
 - Selective
 - Inexpensive
- Modified Support Chemistry Should Affect Catalyst
 - Particle Size
 - Activity
 - Selectivity
 - Life



ELECTRONICS TECHNOLOGY and DEVICES LABORATORY

**US ARMY
LABORATORY COMMAND**

How is a Gas Plasma Formed?

Low Pressure Gas

Apply RF Energy

Gas Dissociates into Ions, Electrons and Free Radicals

These Species React with Available Surfaces

- *
- *
- *
- *
- *
- *
- *

Weakly adsorbed surface species react with gas plasma, dissociate into volatile by-products

Surface reacts with gas plasma. New reactions occur upon exposure to ambient.

Table 1. Physical/chemical properties of chemically and gas plasma treated carbon blacks and the particle size of platinum deposited on them.

	<u>TREATMENT</u>					
	UNTREATED	<u>CHEMICAL</u>		<u>GAS PLASMA</u>		
		ACID	BASE	CF ₄	O ₂	NH ₃
pH	6.5	3.2	6.2	3.3	4.2	6.5
surface area m ² /g	197	---	---	195	192	190
zeta potential mv	-44.2	---	---	-46.4	-46.1	-43.4
Relative ESR spin intensity	1.0	---	---	1.1	1.3	0.7
Pt particle size, Å	74	72	61	59	74	69

Pt and Pt-Ru CATALYSTS

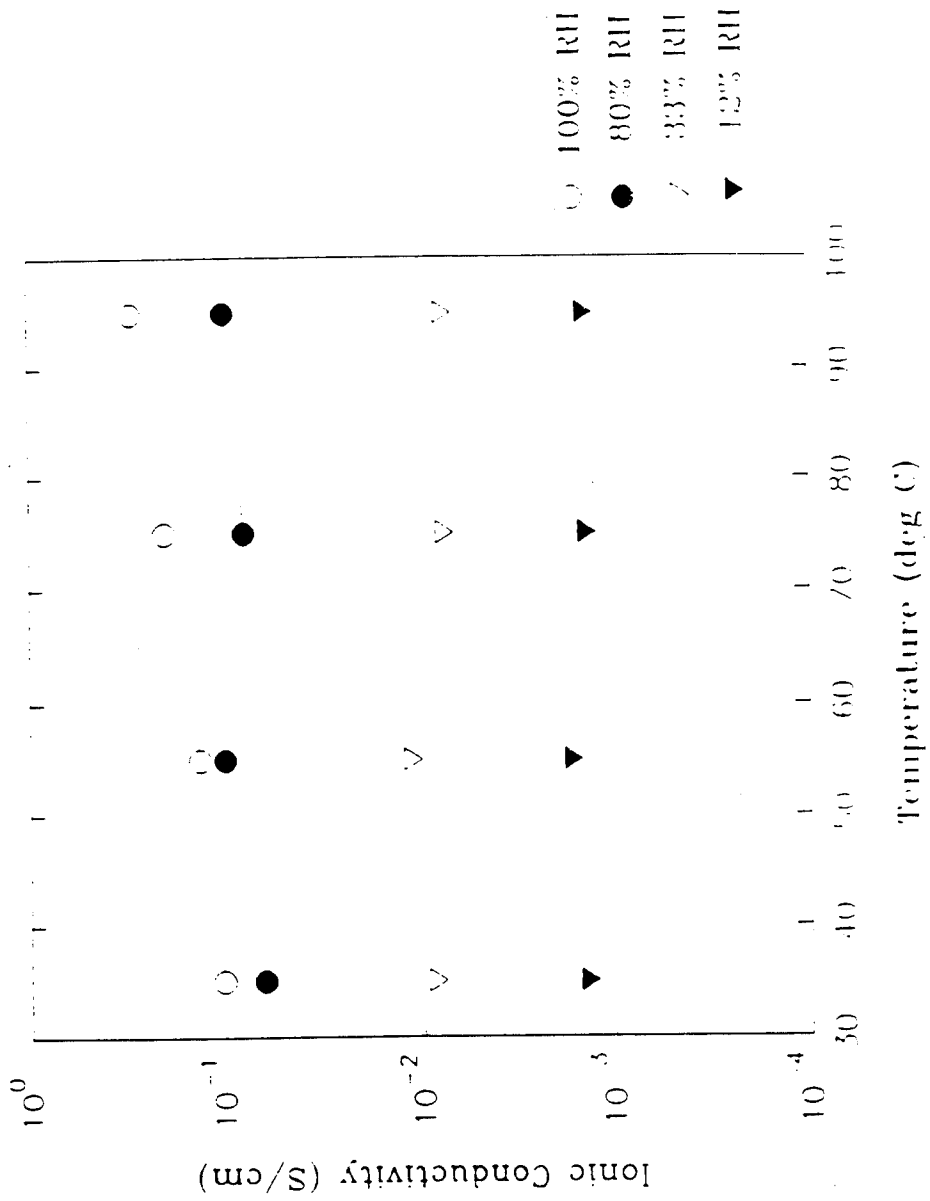
- Dr. Deryn Chu, NRC Postdoctoral Fellow
- Study Effect of Methanol on Oxygen Reduction
- Evaluate Preparation Methods for Pt/Ru Alloys
- Investigate Performance of Various Pt-Ru Alloys

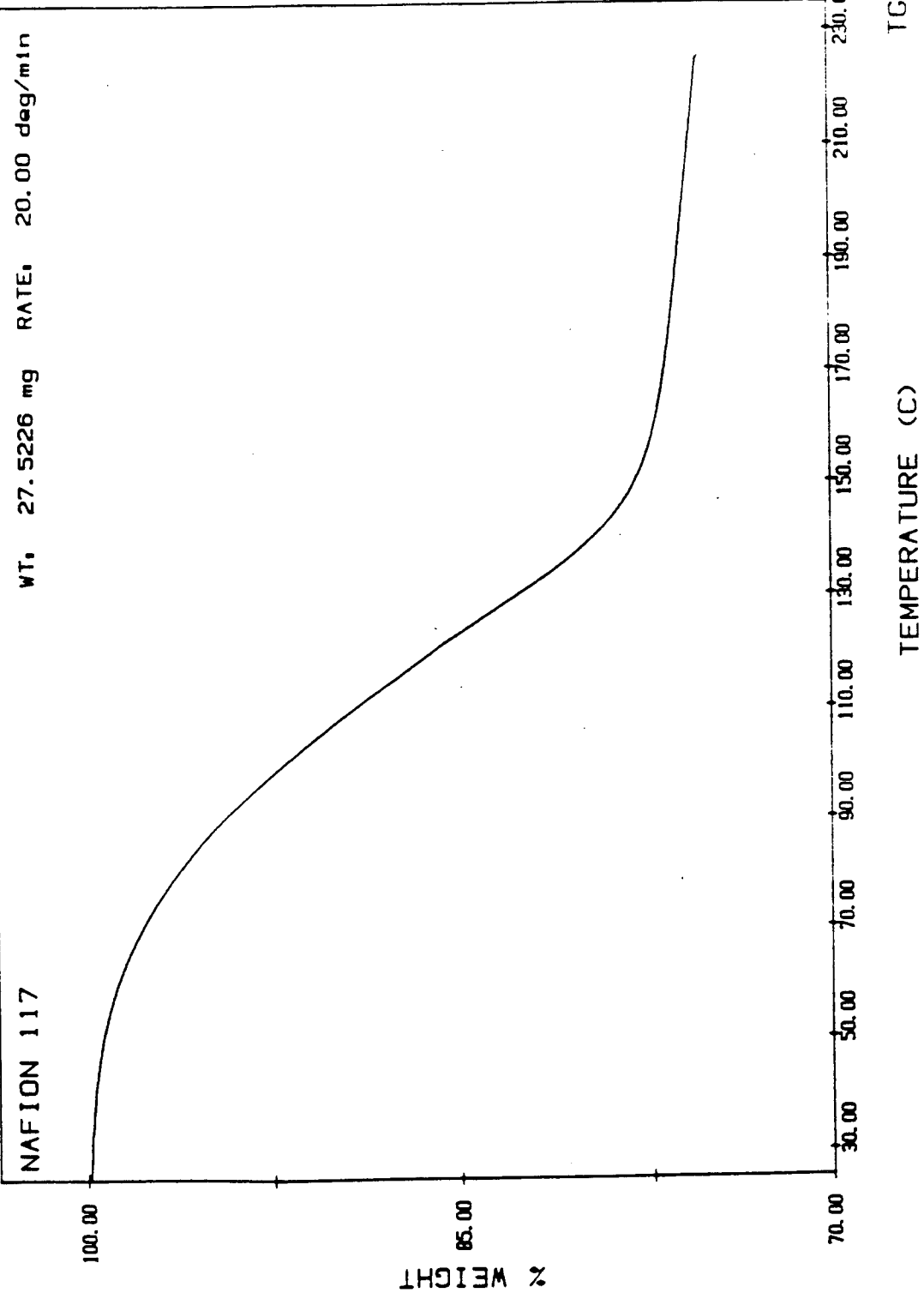


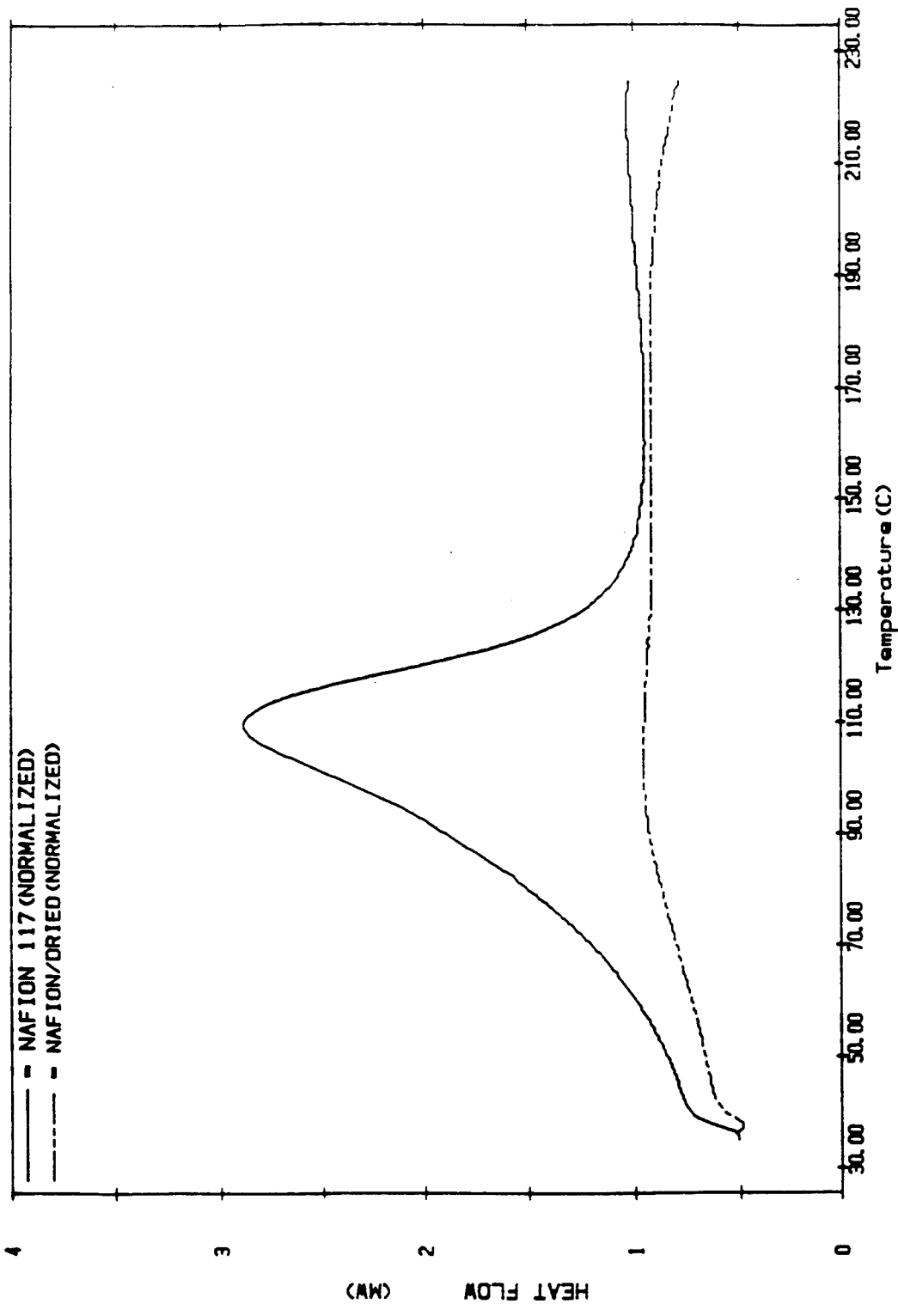
Weight loss of Nafion 117 after equilibrium for 24 hours as a function of relative humidity

Nafion 117	Weight Loss (%)
100% RH	0.38%
80% RH	17.3%
33% RH	23.9%
12% RH	26.3%
dried	28.7%

NAFION 117







DELTA SERIES DSC7

TABLE 1. Endothermic energies of water (KJ/mole) under various conditions.

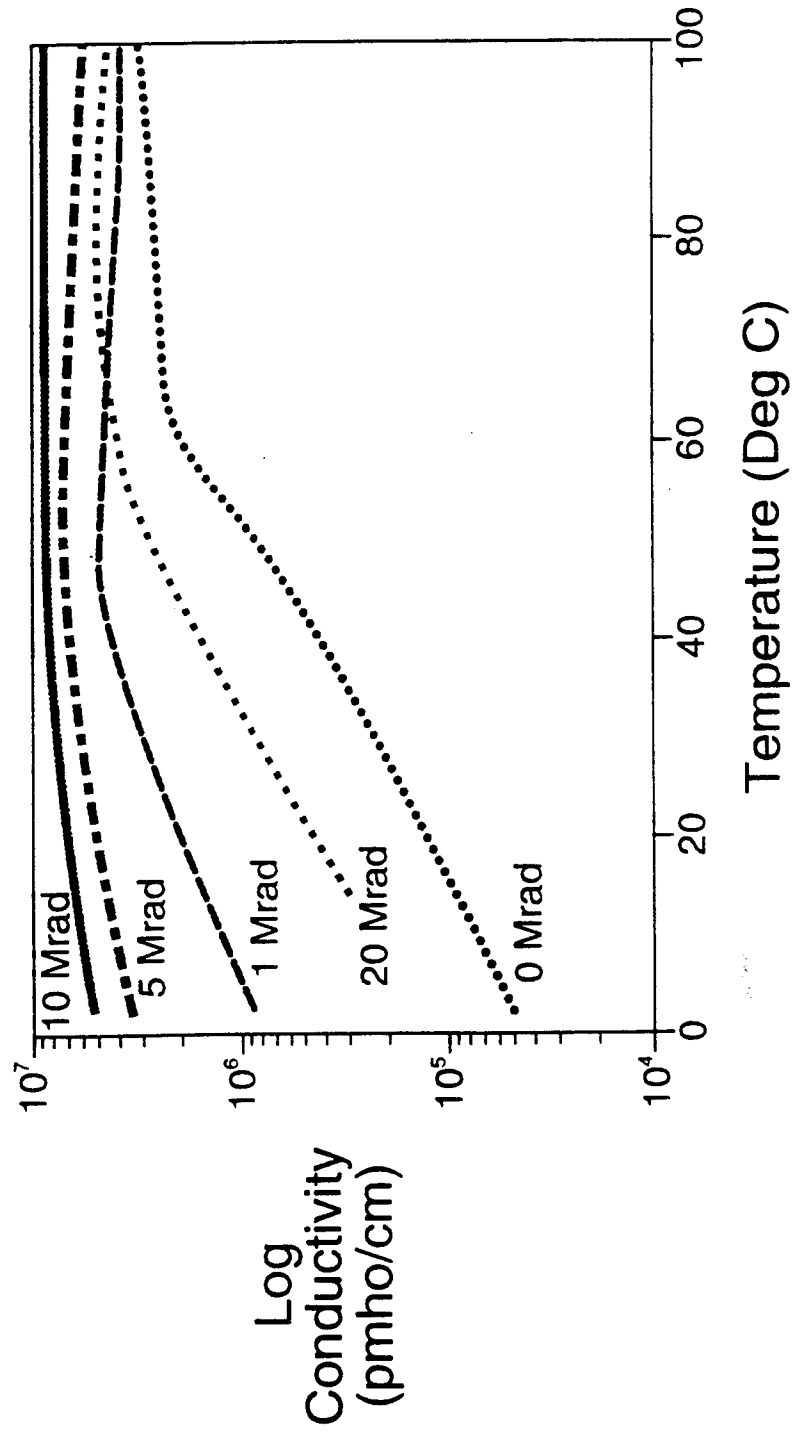
Conditions	KJ/mole
water drop	41.7
in filter paper	39.9
in Nafion 117 membrane	28.9

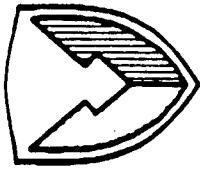
PEM MODIFICATION

Irradiation

- Dupont Corp, Wilmington, DE
R&D Contract
Dr. Shoibal Banerjee/Dr. Warren Buck
- Irradiate Nafion with Electron Beams
Ionic Conductivity
Methanol Permeability

Effect of Irradiation on AC Conductivity of NAFION[®]





US ARMY
RESEARCH LABORATORY

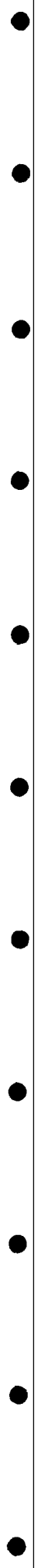
ELECTRONICS and POWER SOURCES

Weight loss of Nafion 117 after drying at 100°C under dynamic vacuum for 24 hours as a function of treatment

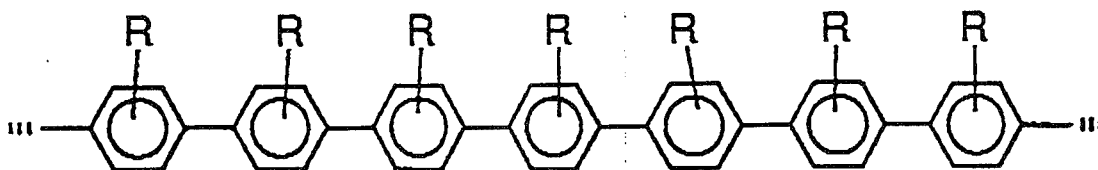
Nafion 117 (dried)	28.7%
Nafion 117 (0.5 MRads)	32.8%
Nafion 117 (2.0 MRads)	35.6%
Nafion 117 (250W/16min NH ₃)	29.6%
Nafion 117 (250W/16min CF ₄)	29.2%

PEM SYNTHESIS POLYPHENYLENES

- Maxdem Corp, San Dimas, CA
Phase I SBIR
Dr. Ying Wang/Dr. Matthew Marrocco
- Prepare New Proton Conducting Polyphenylenes
 - High Ionic Conductivity
 - Low Methanol Permeability
 - Higher Operating Temperatures

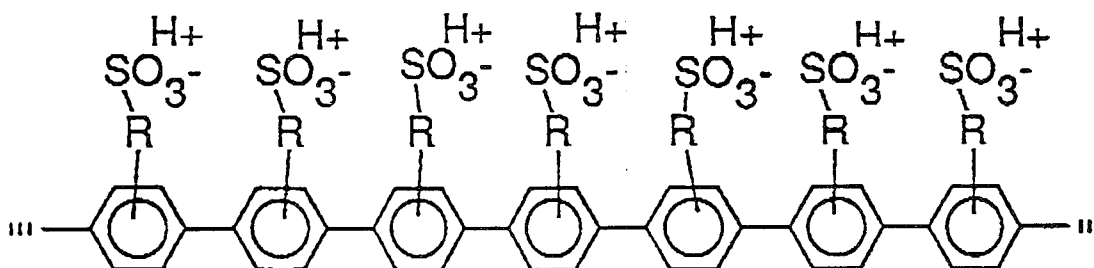


RIGID-ROD POLYPHENYLENES HAVE UNUSUAL MECHANICAL PROPERTIES



- TENSILE MODULUS 1.5 MSI
- TENSILE STRENGTH 35 KSI
- HARDNESS ROCKWELL K 98

IONIC RIGID-RODS MIGHT HAVE UNUSUAL TRANSPORT PROPERTIES



- HIGH IONIC CONDUCTIVITY
- LOW FUEL PERMEABILITY

Maxdem Incorporated

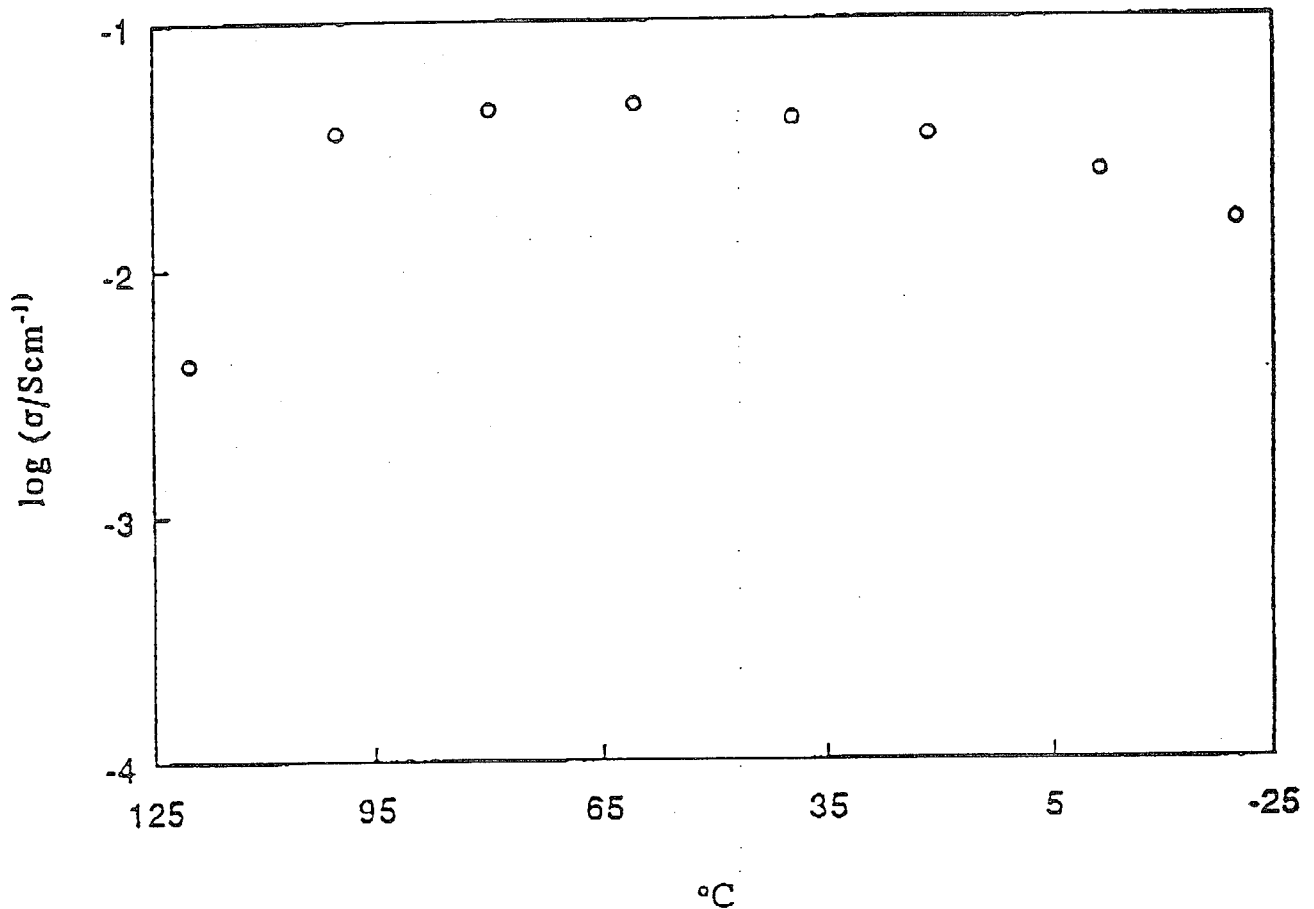
"Polyphenylene SPEs"

10/10/84 10:50 2508 384 0010

MAXDEM INC. 384 00017

CONCLUSIONS:

**FOUND ONE NEW SPE WITH EXCELLENT PROTON
CONDUCTIVITY**



**MUST NOW DEVELOP STRUCTURE PROPERTY
RELATION AND TEST REAL FUEL CELLS**

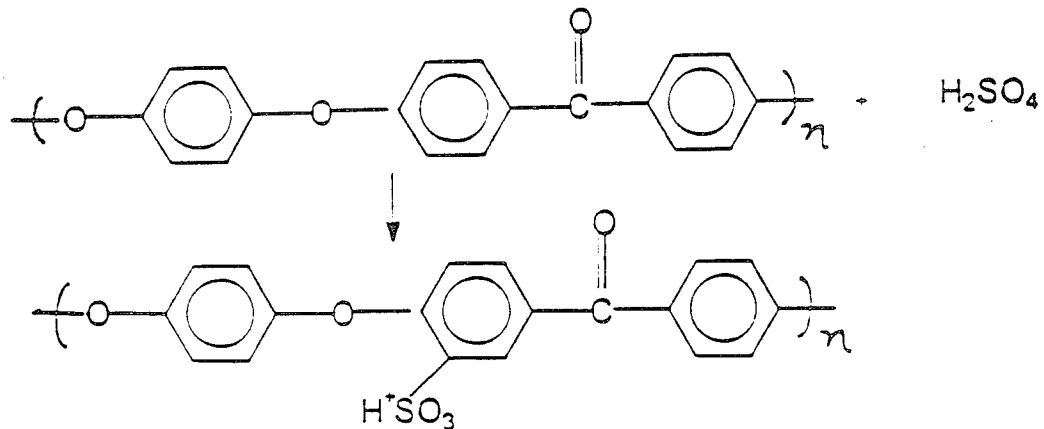
OBJECTIVES

1. Develop new polyelectrolyte materials with low methanol permeability. The new materials will be used as membranes and binders for direct oxidation methanol fuel cell application.
2. Synthesize, characterize and optimize the membrane and binder materials in the areas of film strength, ionic conductivity, methanol permeability and water uptake.
3. Demonstrate a composite membrane for direct methanol fuel cell application.

TECHNICAL APPROACH

1. Proton conducting membrane:

a. The materials used in the new membrane efforts are the sulfonated products of polyether-ether-ketone (PEEK). PEEK is widely used high performance engineering polymer, available at a reasonable price. Sulfonation mechanism shown below.



The conductivity depends on the degree of sulfonation. The degree of sulfonation is time and temperature dependent.

b. The mechanical properties, dimensional stability and methanol permeability of the films are dependent on the degree of crystallinity and degree of crosslinking. Mechanism of crosslinking reaction shown below:

RESEARCH PROGRESS

1. Feasibility study of H-SPEEK as a candidate for a proton conducting membrane
 - a. Sulfonation of PEEK was accomplished.
 - b. Crosslinked films were prepared from the samples that were soluble in acetone.
 - c. The conductivities of the samples were measured by Dr. S Naryanan. Preliminary results are listed below.
 - conductivity comparable to commercial Nafion
 - highly crosslinked films:
 - good mechanical properties
 - high temperature stability
 - low conductivity
 - stable in pure methanol and methanol/water solution

Fuel Cell Research Plans at ARL

where is the money ??

- PEM
 - Structural Modification
 - Composites with gels (water retention)
 - Composites with polymers (higher conductivity)
 - Surface Modification
 - Gas Plasma/U.V. Ozone Treatment to enhance adhesion, reduce methanol permeation, better management
 - Novel Fuel cell Configurations
- Catalyst
 - Study ternary catalysts
 - Alternative substrates (polymers)

Aluminum/Air Semi-fuel Cells for Portable Power Applications

Dr. Bhaskara M.L. Rao, Consulting Scientist
Explorex Inc., 6 Finchley Court, Southampton, NJ 08088.

Aluminum is an abundant, high capacity (3Ah/g, 8Ah/ml), electrochemical fuel. Its use with an air cathode (gas-diffusion electrode) results in a high energy density, Aluminum/Air (Al/Air) semi-fuel cell. The purpose of this paper is to review the recent advances in this field and to show its potential for the development of man-portable power source.

New, high efficiency aluminum alloys and low cost air cathodes have been developed. The cell container is a shell fabricated with the air cathode. It serves as the electrolyte container and holds the aluminum anode in parallel plate configuration with the air cathode of the shell walls. This simple cell design allows easy up- or down-scaling of cell current (A) and capacity (Ah) by varying the area and thickness of the aluminum anode. It can be transported as a dry, light weight and low volume collapsed cell stack (~500 Wh/kg, ~500 Wh/l). It can be expanded and activated with water or electrolyte at the site of operation. Saline or alkaline electrolyte may be used. The salt water version is suited for applications requiring moderate power. The alkaline Al/Air provides high power (>50-100W/kg). Multiple use versions require refurbishing either the aluminum anode and/or electrolyte. These advantages will be discussed with examples of proof-of-concept demonstration.

ALUMINUM-AIR SEMI-FUEL CELL FOR PORTABLE POWER APPLICATIONS

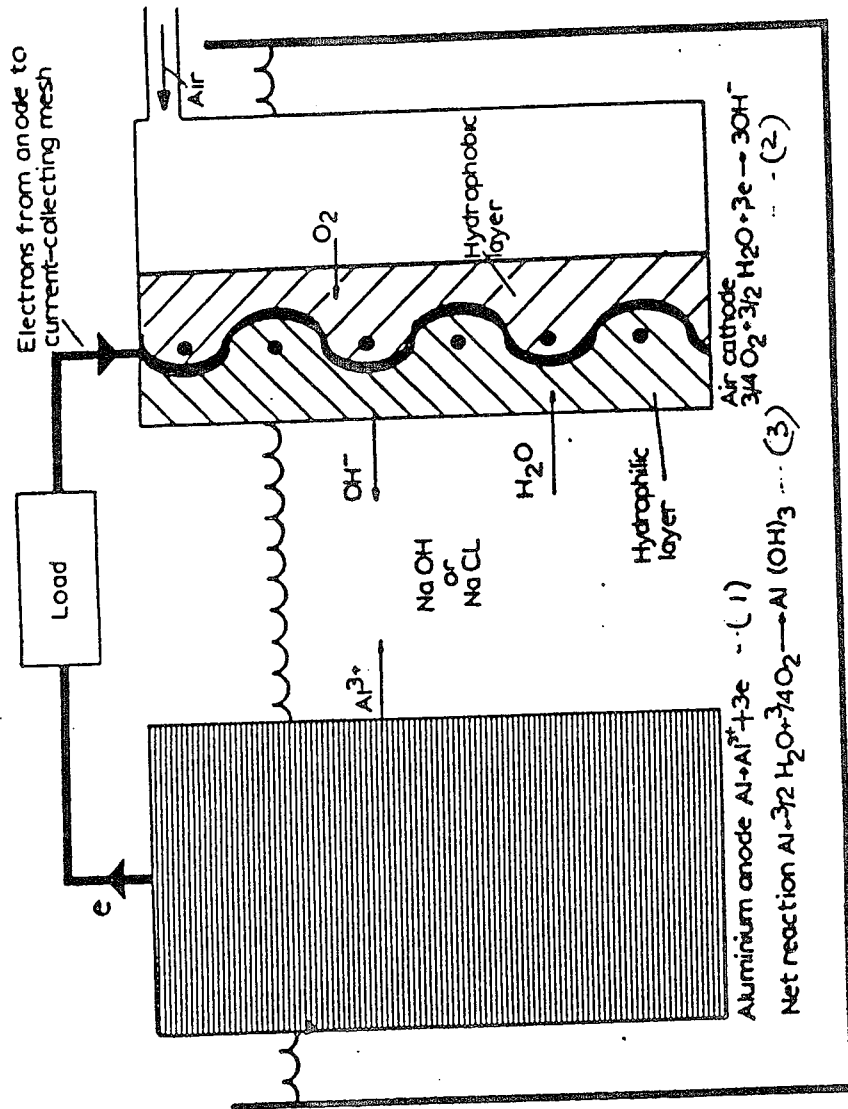
**Presentation at Prospector VII Workshop
October 31 - November 3, 1994**

**BY
Dr. BHASKARA M.L. RAO
EXPLOREX INC.
6 FINCHLEY COURT, SOUTHAMPTON, NJ 08088**

**FOR
YARDNEY TECHNICAL PRODUCTS INC.
82 MECHANICS STREET, PAWCATUCK, CT 06379**

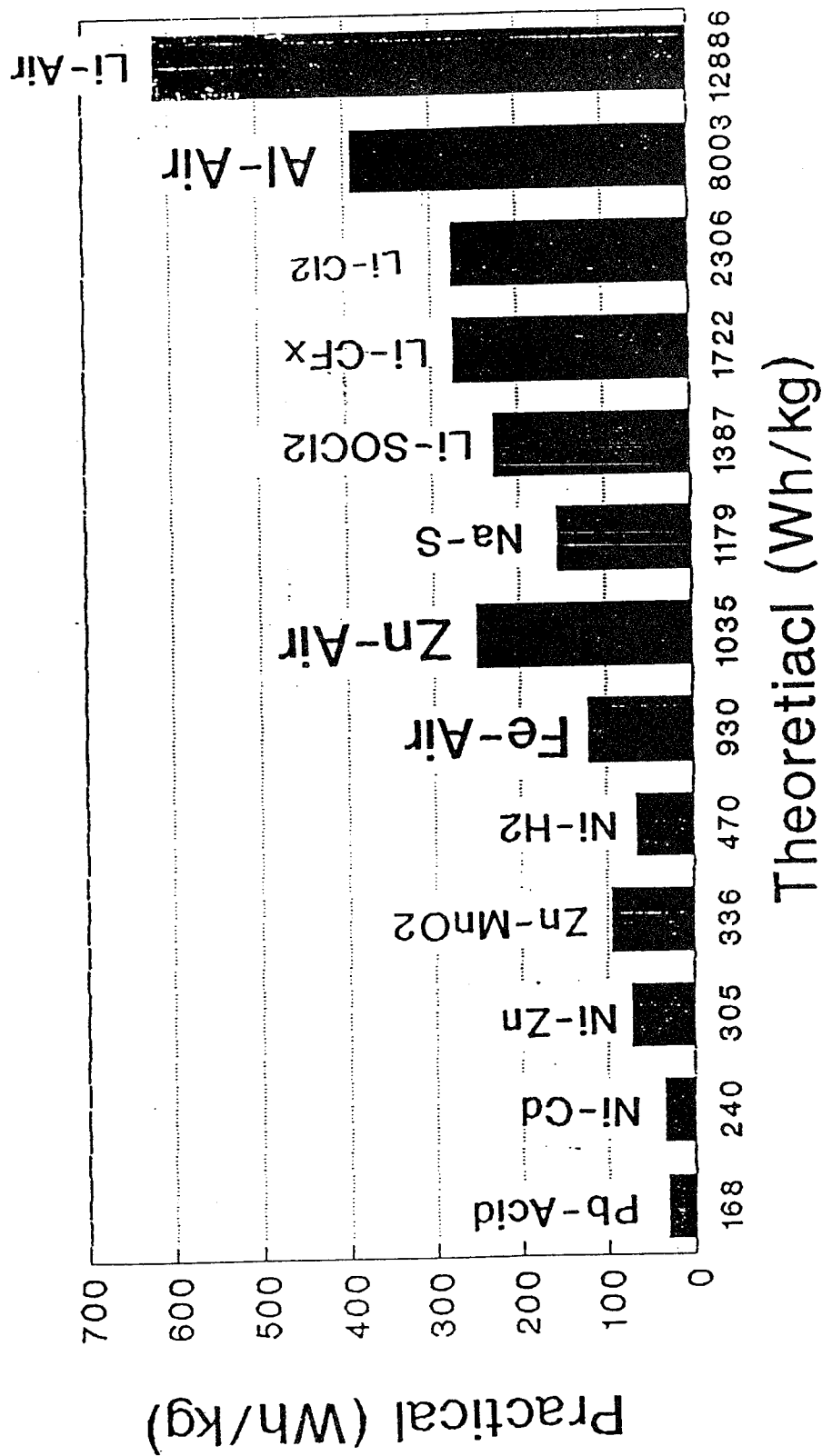
FORMAT OF PRESENTATION:

- **Al/Air Cell**
- **History & Materials**
- **Man-Portable Modes of Application**
- **Proof-of-Concept Discussion**
 - ▶ **NSWC- SOFLAM Module**
 - ▶ **USSOCM- SBIR, Phase-1**
 - ▶ **Re-usable Field Recharger**
- **Summary & Conclusions**

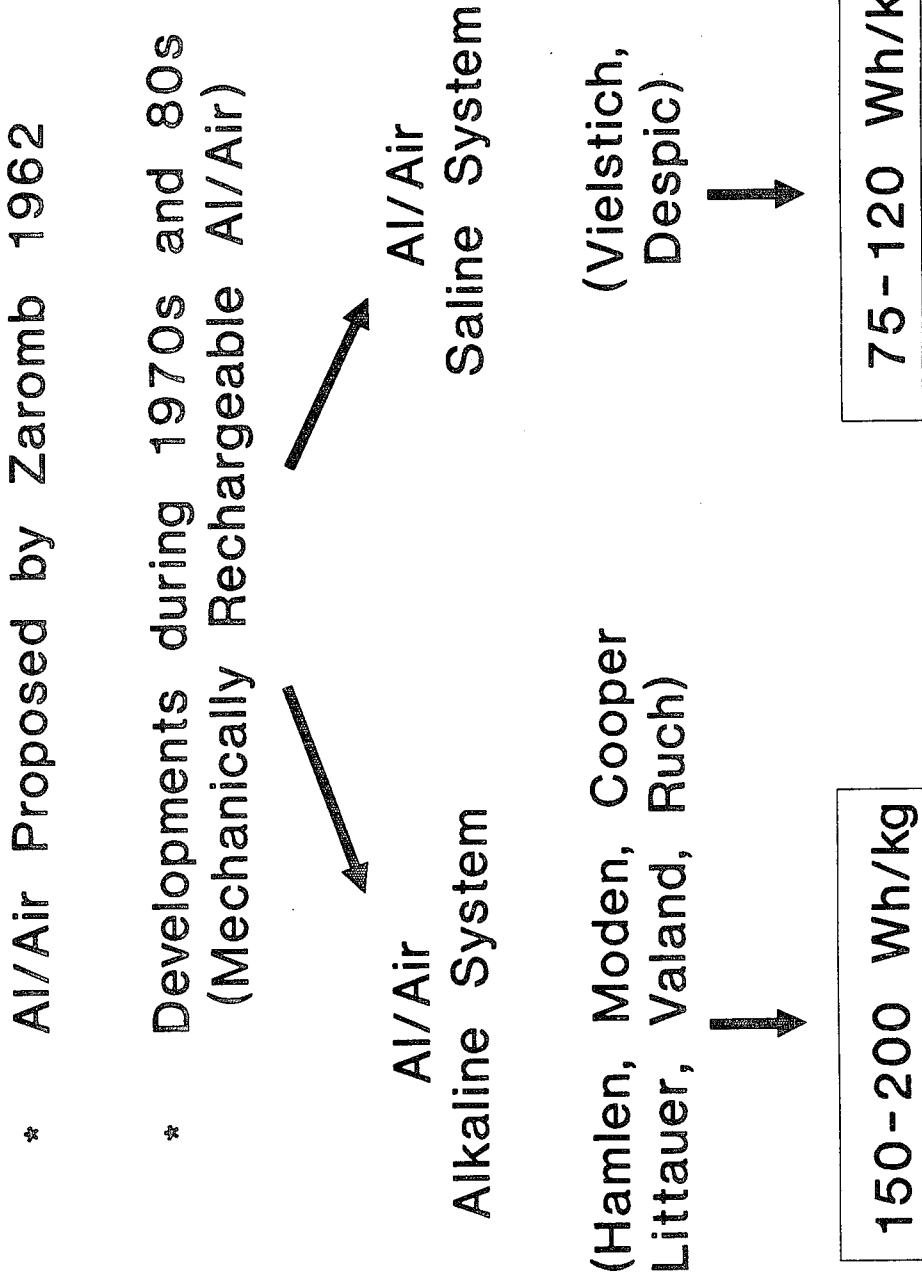


Schematic Operation of an Aluminum-Air Battery

THEORETICAL vs PRACTICAL ENERGY DENSITY



History of Al/Air Technology



RECENT ADVANCES IN MATERIALS AND COMPONENTS

- **High Efficiency Aluminum Anode**
- **Low Cost Air Cathode**
- **New Saline Electrolyte for Re-usable Cell**
- **Solids-free alkaline cell**

DESIGN FLEXIBILITY FOR MAN-PORTABLE APPLICATION

- Collapsibility of air-gap and/or Electrolyte Gap For Reducing Storage & Transport Volume
 - ▶ Uses Air Electrode Shell as Electrolyte Container
 - ▶ Uses Shell with Air Electrode Mounted in Flexible Membrane as Electrolyte Container
 - ▶ Uses Collapsible Spacer

- Dry Storage For Reducing Transport Weight & Increasing Storage Life
 - ▶ Dry Salt or Alkali is Stored in Cell or in Separate Container
 - ▶ Add Water or Electrolyte prepared using local water for on-site Activation
 - ▶ Dump used Electrolyte & Transport, Re-fill at Next Site for Re-use

- Saline or Alkaline Electrolyte Option For Matching Mission Power & Re-usability Requirements

Al/AIR SOFLAM MODULE (NSWC, Crane, IN.)

Requirements

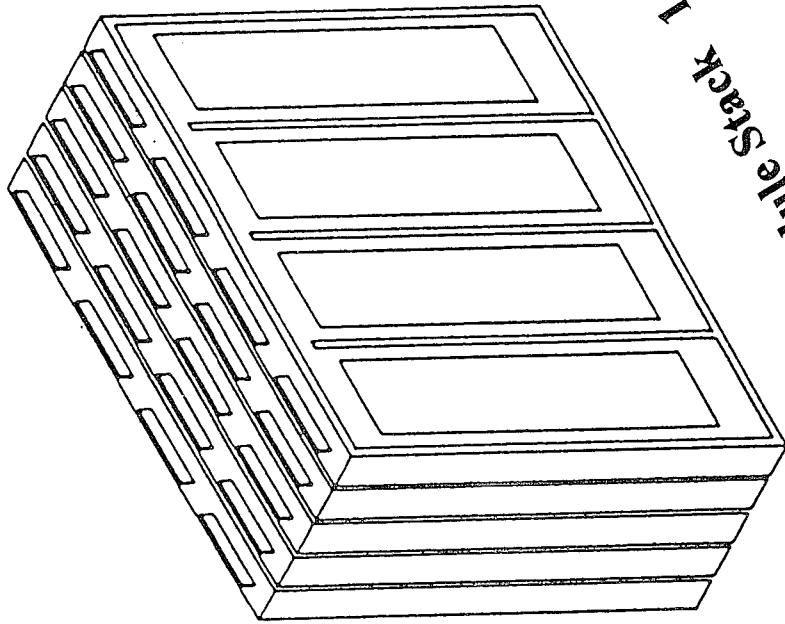
- **Man-Portable**
- **10 Amps, 20-28V, 5-Min on/ 1-Min off
3 Pulses over 15 Min.**
- **Reduced Wt. & Vol. over
Four standard BA5590 (9 lbs, 3.6 liters)**

Al/Air Conceptual Design

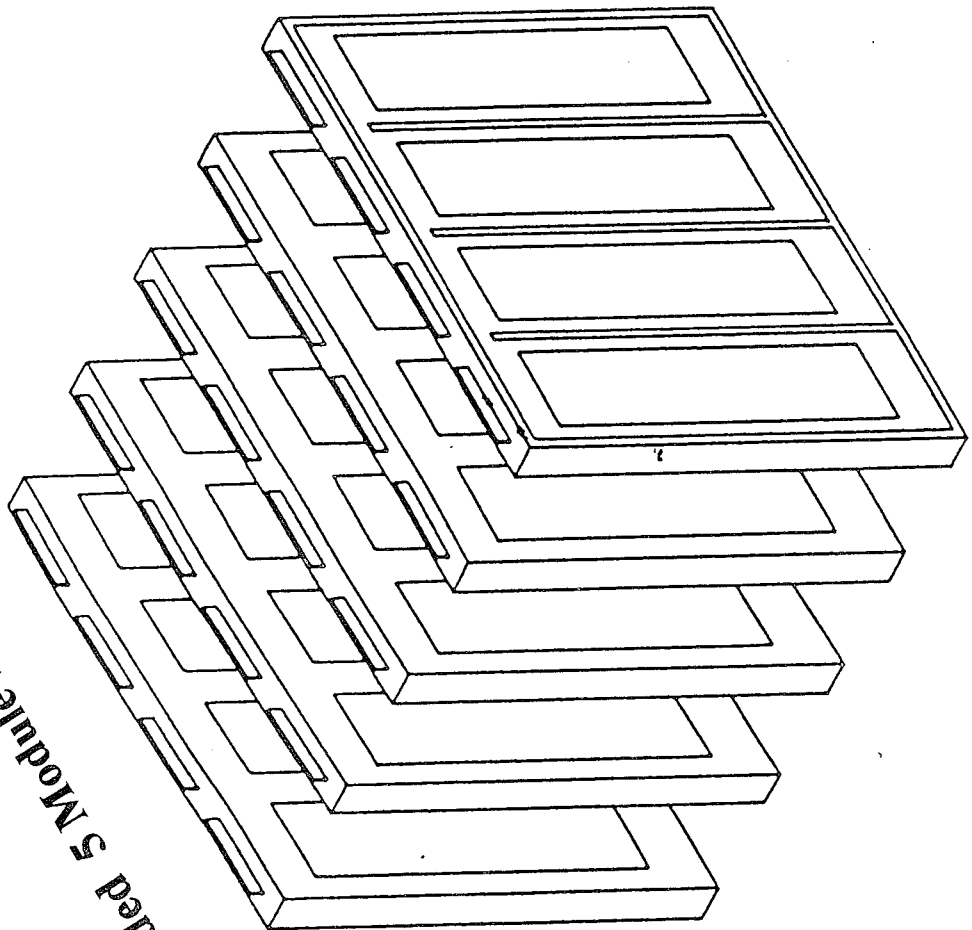
- ▶ **Modular Design**
 - 4 Cells in each module (6V OCV)**
 - 5 Modules in series (30V OCV)**
- ▶ **Dry Stored Collapsible Modules**
- ▶ **On-site Expansion and Activation**

Proof-of-Concept

- **4 Cell module demonstrated**

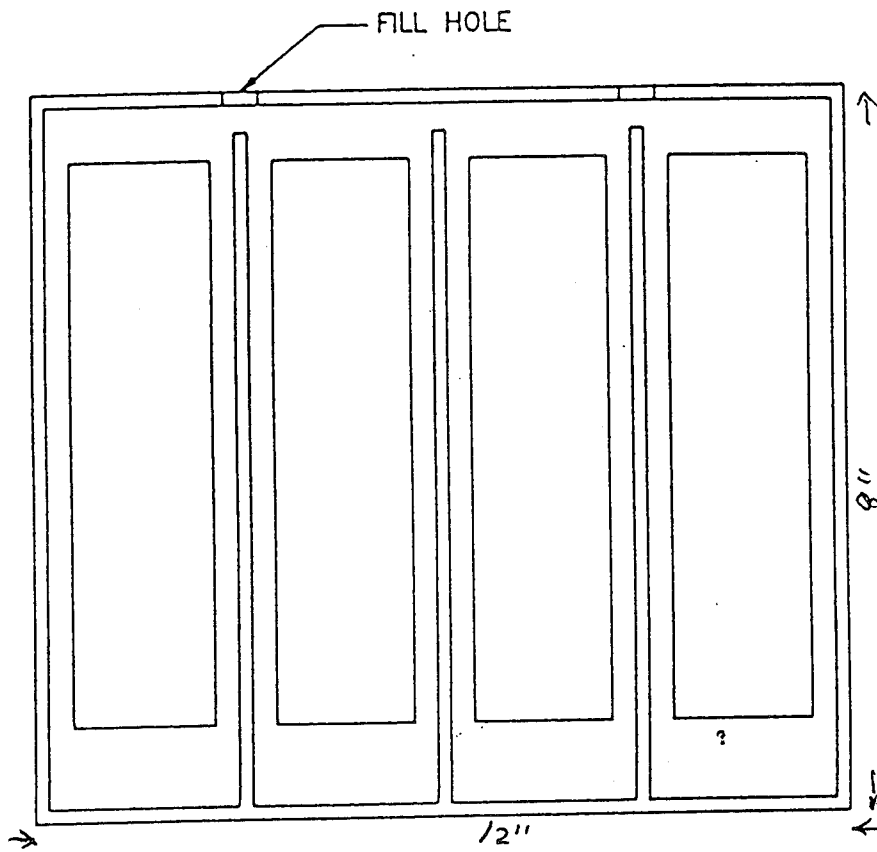
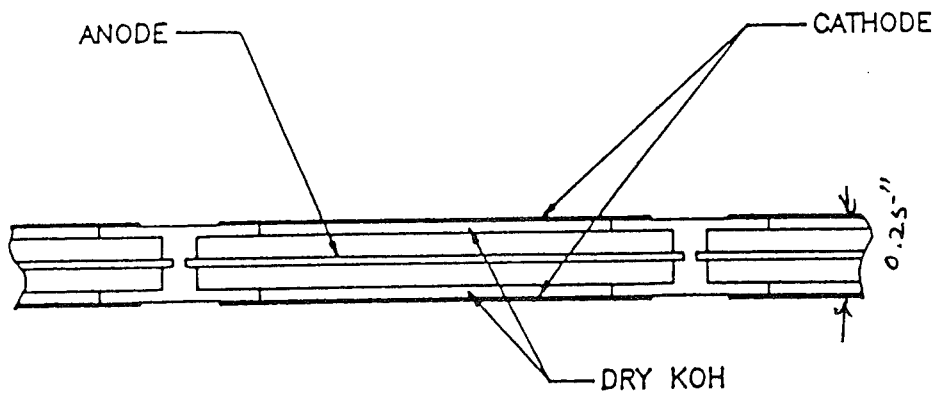


Collapsed 5 Module Stack 4" x 12" x 8"



Expanded 5 Module Stack 4" x 12" x 8"

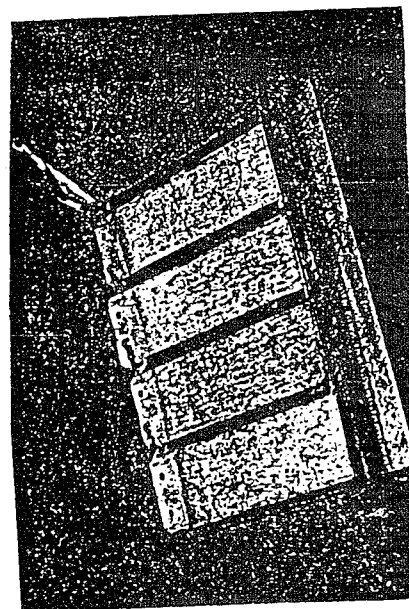
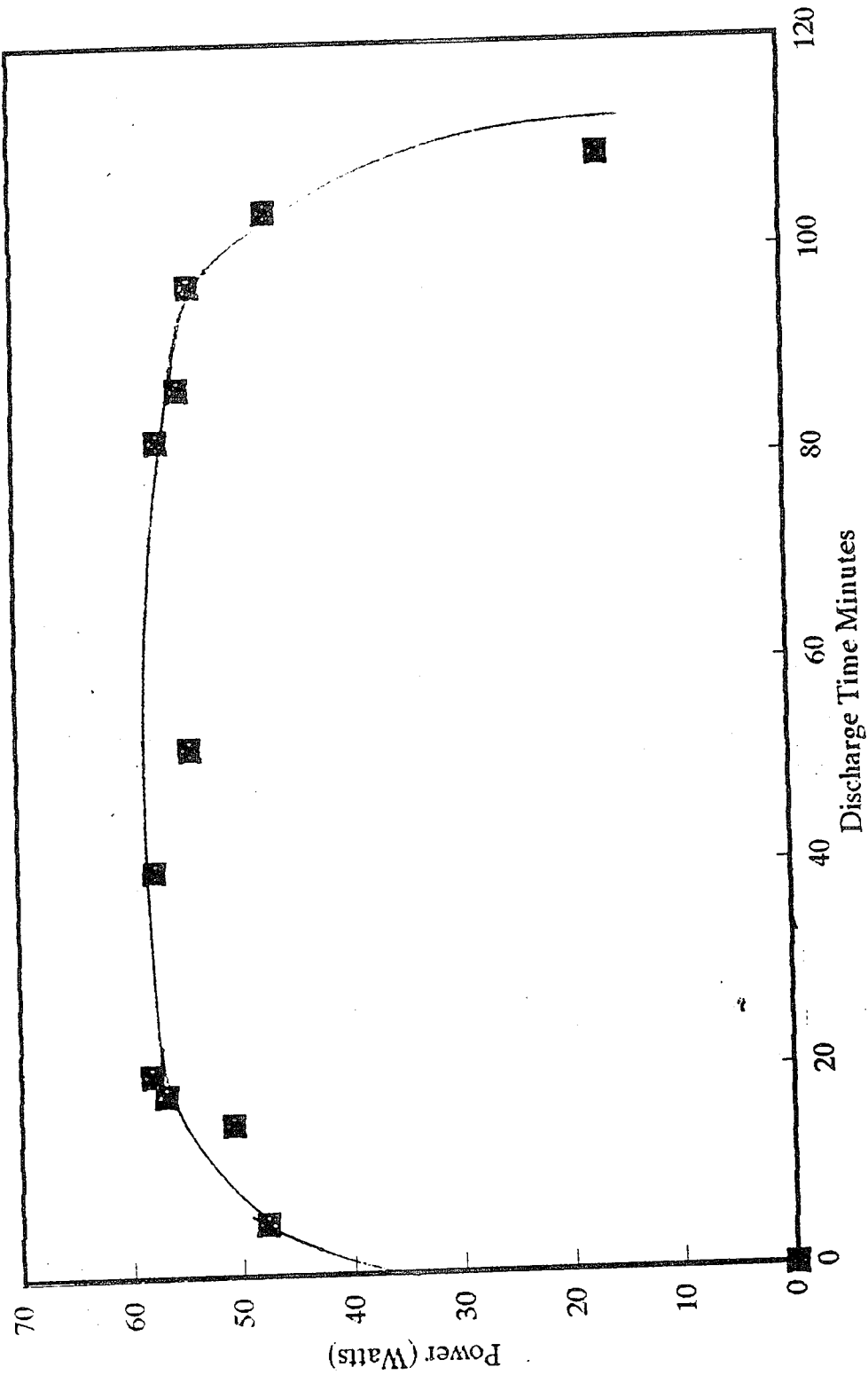
Single Cell 0.25" x 3" x 8"



Four Cell Module 0.25" x 12" x 8"

Four Cell Al/Air Battery Module

10 Amp Continuous Discharge in 8M KOH



COMPARISON OF PROJECTED WEIGHT AND VOLUME

Power Source	Weight (kg)	Volume (Liters)
Al/Air (Dry, Collapsed)	1.4	2.4
4 BA5590	4.2	3.6

Al/Air INVESTIGATION FOR SOCOM-93-001/SBIR, Phase-1 (Commonwealth Tech. Inc.)

OBJECTIVE

**ADVANCED DEVELOPMENT OF Al/AIR BATTERY FOR
COMMUNICATION EQUIPMENT**

Phase-1 Develop & Document Battery Design

REQUIREMENT

- **Majority of SOF Communication Equipment
Require 2.5 to 3.0 Amps at > 22 Volts.**
- **Mission lasts 7 to 10 Days**

Al/AIR DESIGN

- ▶ **3.6 Amps at > 22 Volts to
Power at 14 Volts or 28 Volts**
- ▶ **Plug Compatible with BA5590/U,
BB590/U and BB490/U**
- ▶ **Rated 3.6 Amps >25 Volts for 35 Hours**
- ▶ **Al/Air or Al/Air-NiCd Hybrid**

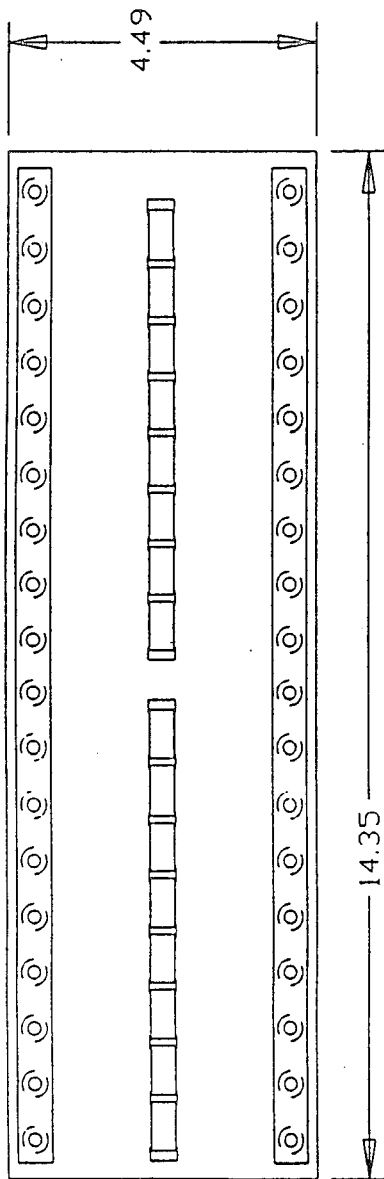
DESIGN MODELING RESULTS

- **One-shot, Alkaline System Preferred
To Optimize Weight, Volume and Power**
- **Fixed Volume Satck**
- **Two Design options Examined**

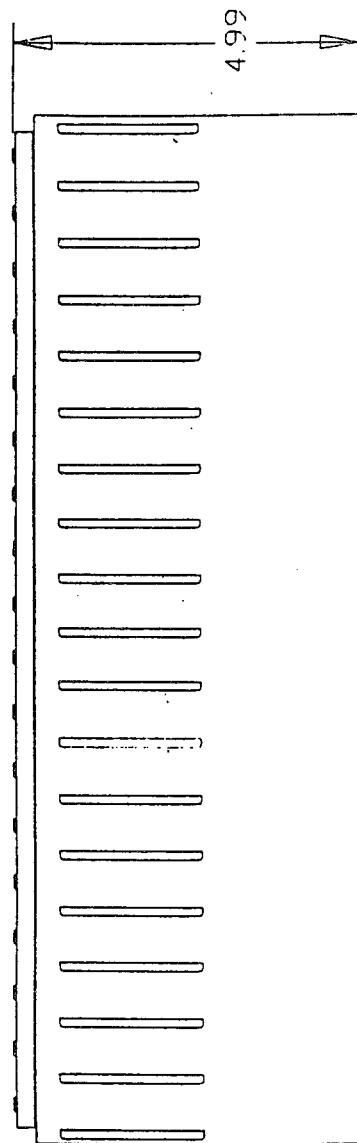
1. 18 Cell Al/Air	3.2 Kwh
4.5" x 5" x 14.4"	5.3 liters
2.. 16 Cell Al/Air - NiCd Hybrid	2.8 Kwh
4.5" x 5" x 15.1"	5.5 liters
- **Hybrid Favored to Accommodate Higher Power
Requirement For Satellite Communication**

SCHEMATIC OF 18 CELL AIR/AIR POWER SOURCE

Top View



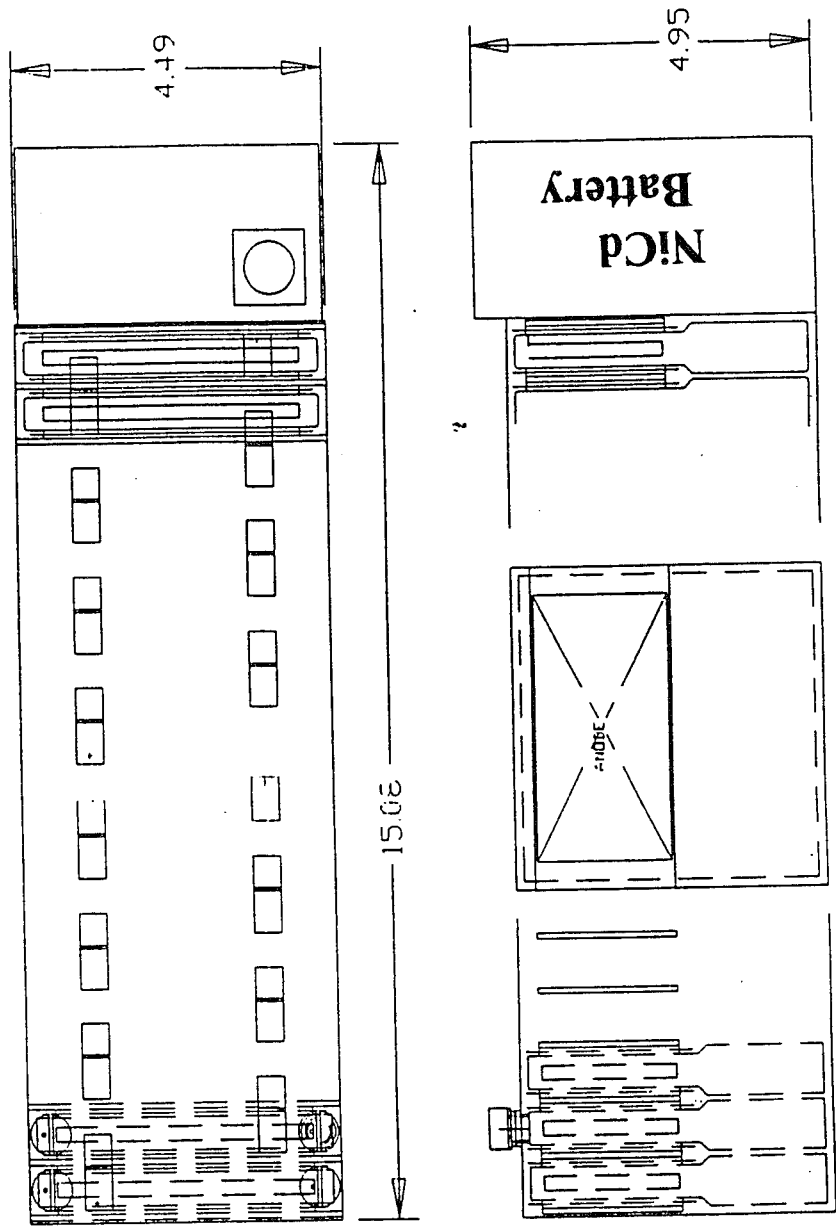
Side View



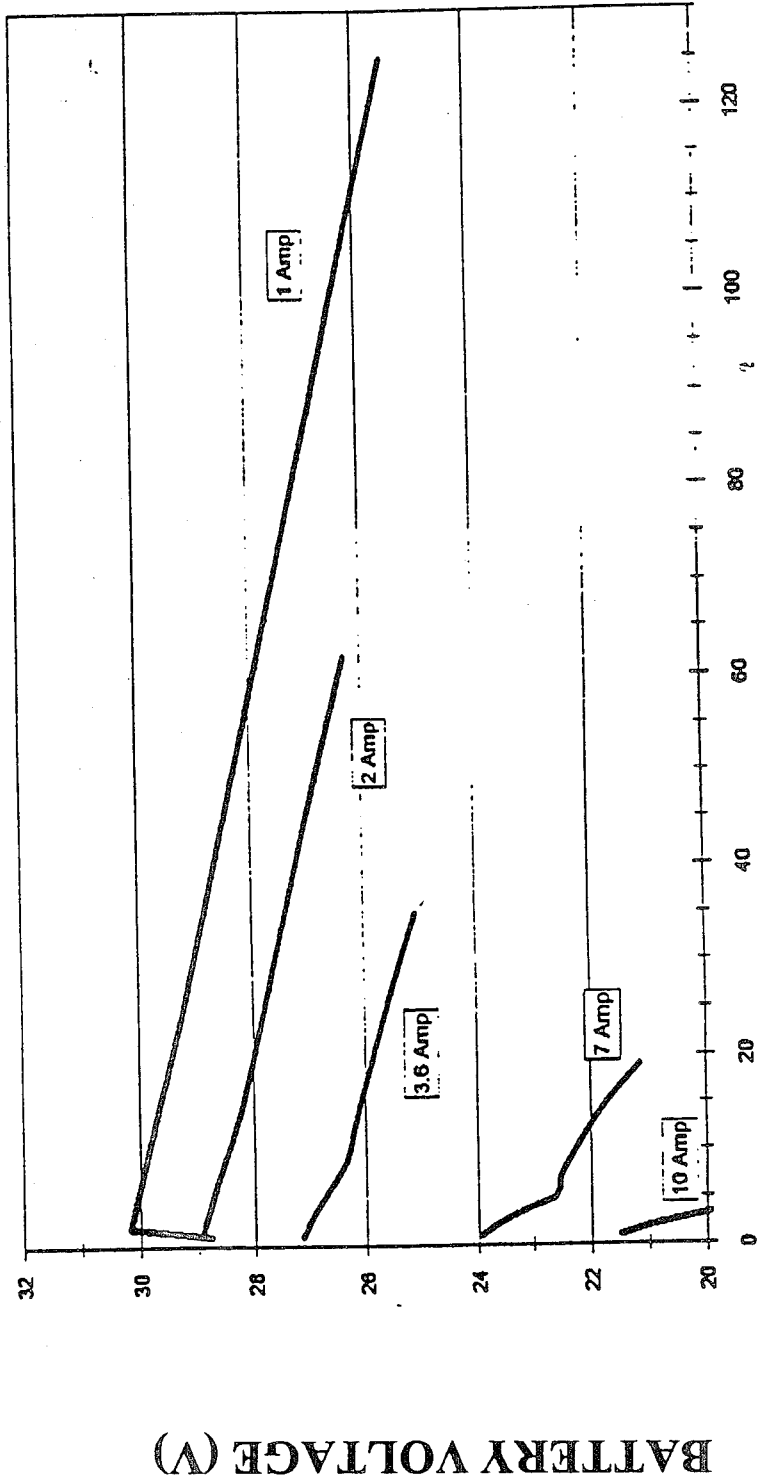
**Active
Region**

**Precipitate
Tank**

SCHEMATIC OF 16 CELL A/AIR - NiCd HYBRID SYSTEM



PROJECTED PERFORMANCE OF 18 CELL A/AIR BATTERY



DISCHARGE TIME (HOURS)

BATTERY VOLTAGE (V)

COMPARISON OF 3.2 kWh Al/AIR WITH BB5590 LITHIUM BATTERY

Battery	Energy Per Unit (Wh)	# of Units to Match 3.2 kWh Al/Air
BA5590/U	144	22

Comparison on Weight Basis Al/Air Weight: ~3 kg (Dry wt. Stack+salt)

Battery	Weight (kg) for 3.2 kWh	Al/Air Figure-of-Merit
BA5590/U	23.3	7

Comparison on Volume Basis Al/Air Volume: ~6 liters (Stack+salt)

Battery	Volume (l) for 3.2 kWh	Al/Air Figure-of- Merit
BA5590/U	19.6	3

AI/AIR SYSTEM FOR FIELD CHARGING NiCd & LEAD-ACID POWER PACKS

Drawbacks of current Technologies

- **Solar Panels**
 - : Weather Dependent
 - : Require Operator's Effort for Regular Repositioning
 - : Risks Exposing Operators's Position
- **Hand Cranked Generators**
 - : Require Rigorous Operator's Effort

Key Requirement for New System:

- Minimize Operator's Effort

AI/Air System (*) Concept

Saline

AI/Air 6A,6V

Battery

DC/DC 34V,0.75A
Converter

NiCd

or

Lead-acid

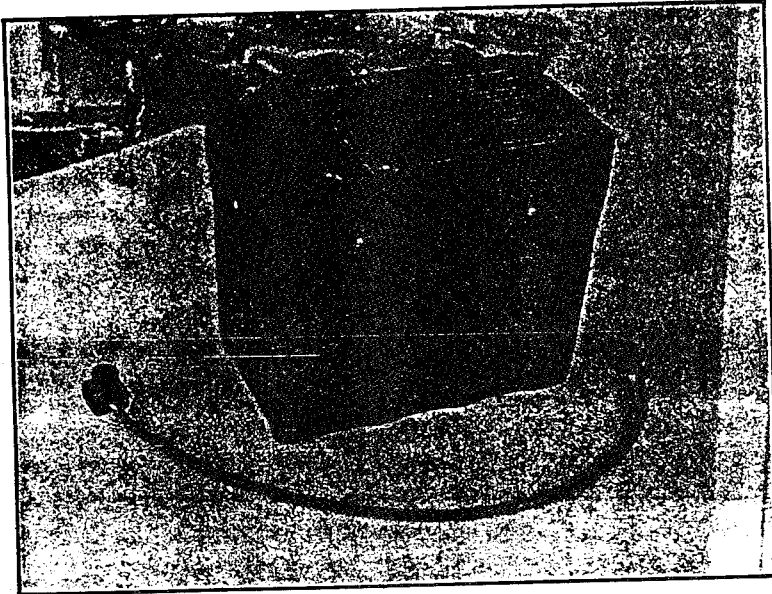
(*) Sponsored by OSD-C31,ST

Ref: S.T. Winarski et al, 36th PSC, June '94, p126-128.

Aluminum-Air Field Recharger

600 Watt-Hours, 6 Volts Secondary Battery Recharger

Recharges a fully discharged BB490, BB590, BB690 in 4 to 5 hours. (7 + cycles)



Advantages

- Silent
- No Maintenance
- Light Weight
- Long Shelf Life
- Water Activated
- Disposable
- Environmentally Friendly

Smart System

- Recharges fast and safely
- Shows charge status
- Automatically switches to maintenance charge mode
- Reusable Power Module
- Detects faulty Battery
- Provides correct charge independently

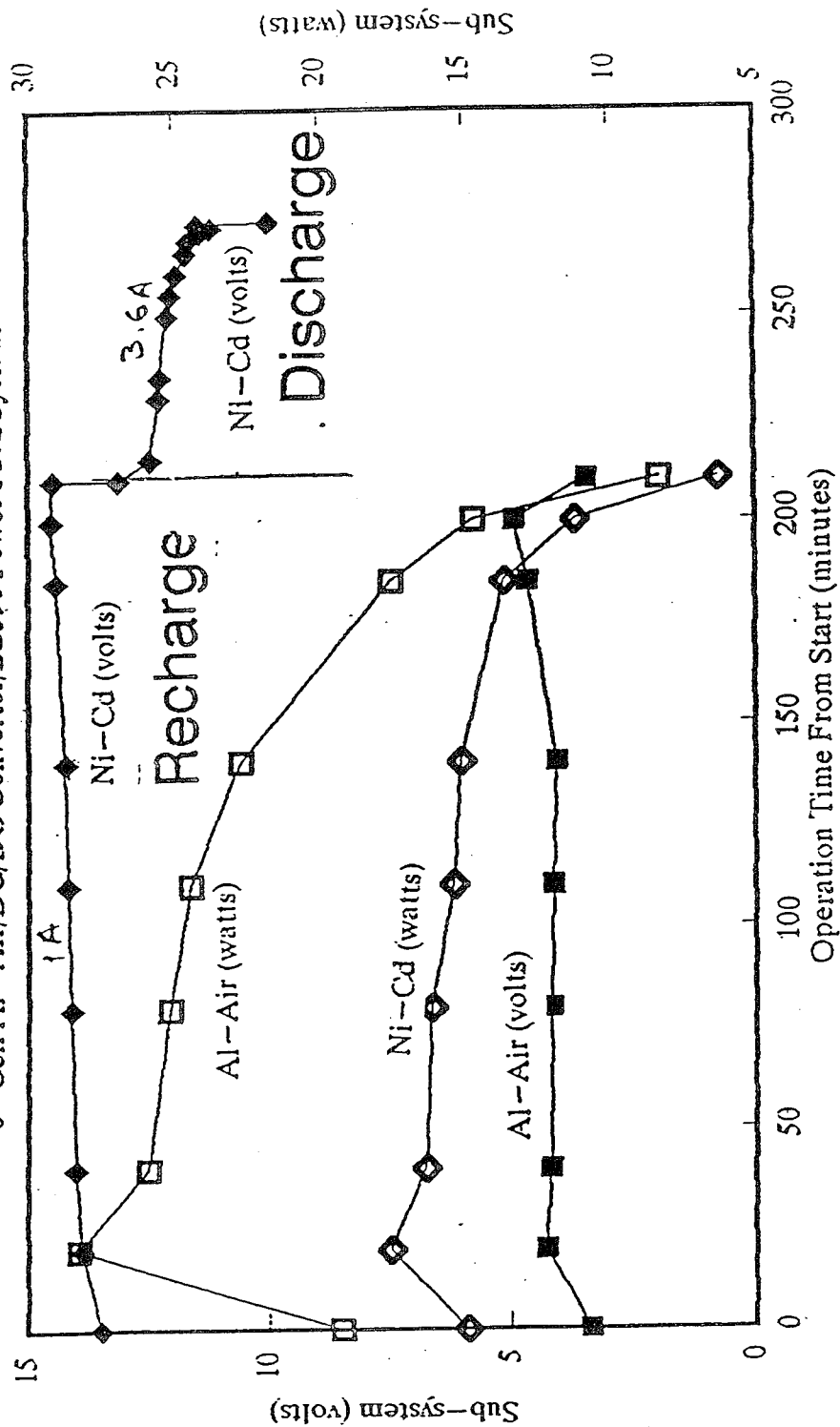
CHARACTERISTICS

BATTERY WEIGHT - 4 lbs. (1.8 kg)
DIMENSIONS - 9" x 5" x 8" (5.2 l)
VOLTS On Charge - 29 V
AMPS On Charge - 650 ma

POWER MODULE - 116 (45g)
SALT BAG - 1/2 lb. (290g) each
ENERGY DENSITY - 294 Wh/kg
102 Wh/l

Third Recharge & Discharge of BB590/U

6-Cell Al-Air/DC/DC Converter/BB590 Power Pack System



150 ml Modified Electrolyte
File0118.wk3/B-26, P134/BR

TEST RESULTS & RECOMMENDATIONS

Govt. Testing at NSWC, Crane, IN.

**Partially & Filly discharged
2 Ah BB590/U, 3 Ah Super NiCd &
1.8 Ah BB490/U Successfully Tested**

USASOC Field Test by Soldiers & Recommendation

Praised performance.

**Recommended packaging the device as
separable smaller modules for ease of packing.**

Evaluation Report statement

**AI/Air Technology is a "substantial improvement" and
it has "extremely promising potential."**

SUMMARY & CONCLUSIONS

- **High Energy Density, recent developments in materials, saline and alkaline electrolyte option are attractive features of Al/Air for the development of Man-portable Power Source**

- **Proof-of-concepts favoring Al/Air based Man-portable Power Source demonstrated :**
 - ▶ **Collapsibility of air-gap and/or electrolyte-gap for reducing storage & transport volume**

 - ▶ **Dry storage for reducing storage & transport, and for long shelf-life**

 - ▶ **Simple fabrication using air electrode shell as electrolyte container**

 - ▶ **On-site activation by electrolyte prepared using local water**

 - ▶ **Re-use by dumping used electrolyte & transport, re-fill at next site**

- **Field test report supports that Al/Air Technology has extremely promising potential development of Man-portable Power Source**

TECHNOLOGY UPDATE II

TECHNOLOGY UPDATE SESSION II

Chairman: Dr. Bob Guenther, ARO

The session began with introductory remarks by the Chairman commenting on the diversity of approaches to be discussed in this session. The topics ranged from an analysis of system integration aspects of fuel cell devices, to modeling of fuel cell systems, to electrochemistry aspects of fuel cell electrode materials and electrochemistry issues of FC systems, to reports on studies made of subsystems for these small portable fuel cells. An interesting report was given on an innovative laminar structure concept (zero crossover direct methanol fuel cell) under development by one researcher.

The lectures presented included the following:

- "System Integration Aspects of Small Portable Devices," Dr. Scott Hynek, Arthur D. Little
- "Energy Storage: Manpack Hydrogen Tanks," Mr. Ira Kuhn, Directed Technologies, Inc
- "Oxygen Storage Systems," Mr. Jerry Chodil, Ball Aerospace
- "Direct Methanol PEM Fuel Cell Development," Dr. Bob Savinell, Case Western Research Institute
- "Evaluation & Modeling of PEM Fuel Cells," Dr. Ron Mann, Royal Military College of Canada
- "Electrodeposition of Nafion on Electrode Structures," Dr. Bruce Tatarchuk, Auburn University
- "Innovative Laminar Structure Concept," Dr. Eugene Smotkin, Illinois Institute of Technology

The remainder of this section is a compilation of the technical materials as submitted to the Workshop organizers.

SYSTEM INTEGRATION ASPECTS
OF SMALL PORTABLE DEVICES

Scott Hynek
Arthur D. Little, Inc.
Cambridge, MA

ADL's specific expertise in portable fuel cell system integration derives from performing an extensive preliminary design for the SNORKLER solicitation. Additionally, we have a substantial background in large system integration, and plenty of experience in designing portability into military systems.

For example, we designed the load-bearing components of the Soldier Integrated Protective Ensemble (SIPE), and are integrating the load-bearing components and the armor of the 21st Century Land Warrior - Generation II. We recently designed a hand-held mass spectrometer for astronauts to use in constructing the space station, to detect leakage and personal contamination. We re-designed the head mount for the Night Vision System, to permit the user to use it while moving.

A systems integrator, like a symphony conductor, should be a generalist. A conductor should be able to play all instruments, though he need not play any of them as well as the musicians he conducts. Similarly, a systems integrator needs to know enough about the design of each component to set reasonable weight, volume, and power consumption goals for each; to select their operating points; to provide for the needs (power, feedstream, discharge, etc.) of each component; and to determine a suitable control architecture for the entire system.

A systems integrator may be called upon, in the course of a design, to define tradeoffs that permit the customer to modify system specifications. For example, once the design has progressed to the point that the system integrator can quantify the relationship between the maximum ambient operating temperature and the operating duration, he should describe that relationship to the customer so that the customer can select the combination of operating temperatures and duration that best suits his needs.

A systems integrator should provide visual context to the customer and to the component suppliers. Both need to see system layout drawings from the very beginning, and both are even better served by a three-dimensional system mockup.

Portable fuel cell power systems present special challenges to the system integrator. In addition to the obvious challenges (low weight, low bulk, ruggedness, reliability, and low noise) there are concerns with respect to

- water management - the process air stream needs water to keep the fuel cell membrane hydrated, the chemical hydride system needs water as a feedstream, the fuel cell makes water as an

inescapable consequence of its operation, and any surplus water cannot be visible if the fuel cell power supply is to operate clandestinely;

- thermal considerations - the chemical hydride system must be cooled, the metal hydride system must be heated, the fuel cell stack can only generate full power once warmed up, and the fuel cell stack must be cooled if the ambient temperature is high;
- modes of operation - storage, transport, standby, startup, low-power, and high-power;
- safety (during transport, setup, and refueling); and
- determining the remaining capacity in each fuel system.

Though the fuel cell stack deserves and gets most of the attention, there is much more to a portable fuel cell power system than the stack alone. The system integrator must package and control the stack along with primary and secondary batteries, process and cooling air fans and ducting, various hydrogen sources, the water reservoir and pump, and the moisture exchanger.

In summary, a portable fuel cell power supply will meet its weight, bulk, and duration goals, and operate safely and reliably, only if the system integrator has done his job well.

System Integration Aspects of Small Portable Devices

*Prospector VII:
Small Fuel Cells for Portable Power*

November 1, 1994

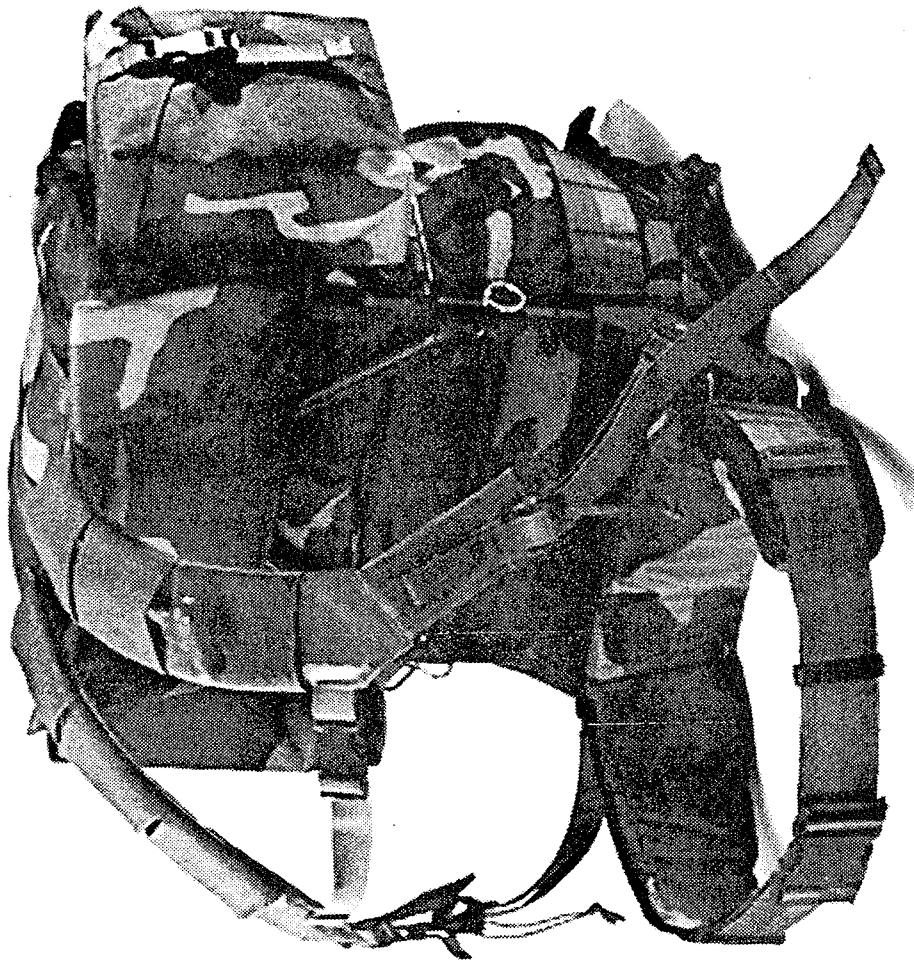
Arthur D Little

Scott Hynek

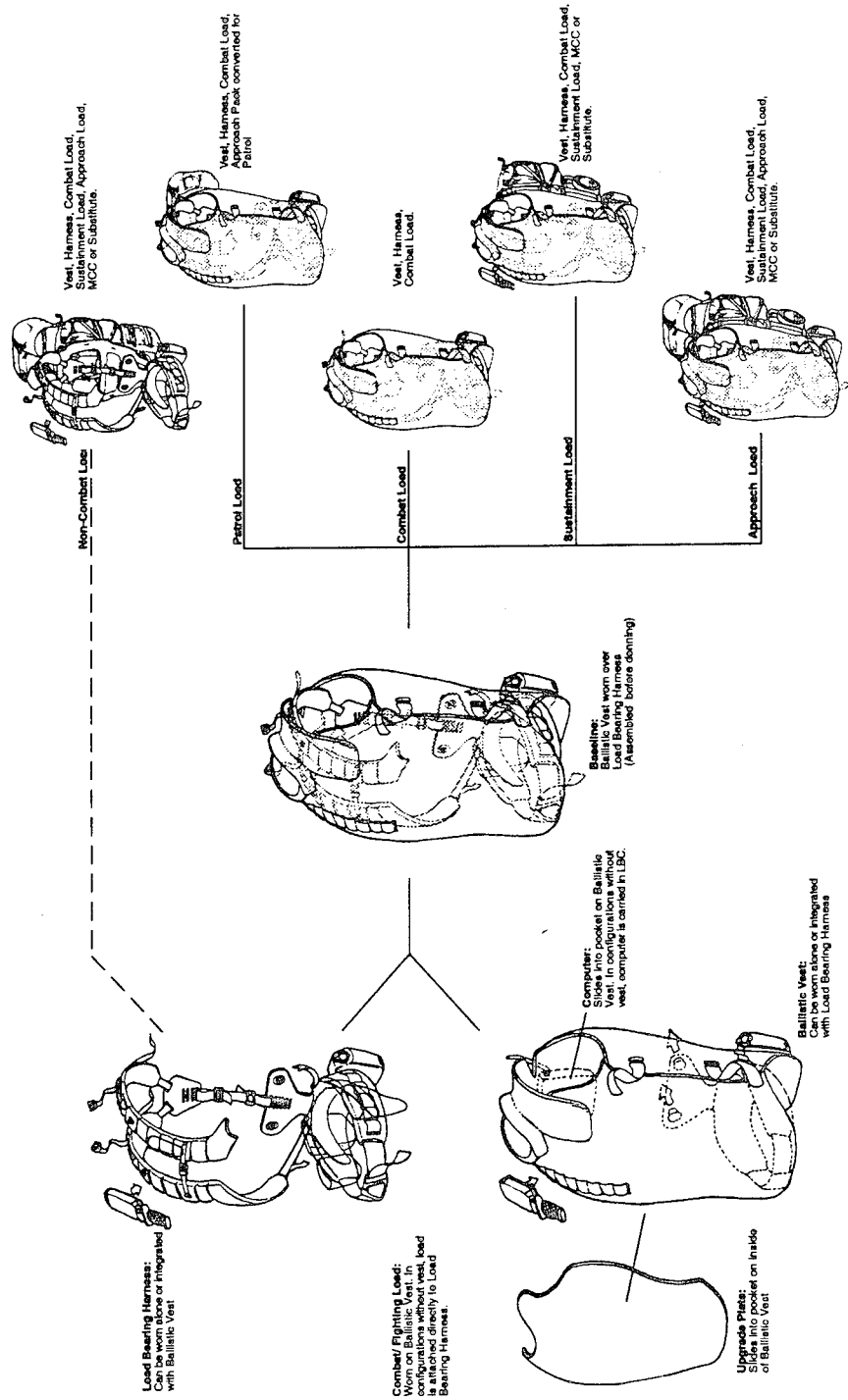
ADL's Experience in Portable Systems Integration:

- ◆ **Power Systems**
 - **SNORKLER Design Study**
- ◆ **Systems That Include Power Systems**
 - **Personal Cooling Systems**
 - **Soldier Integrated Protective Ensemble (SIPE)**
 - **Night Vision System**
 - **Hand-held Mass Spectrometer**

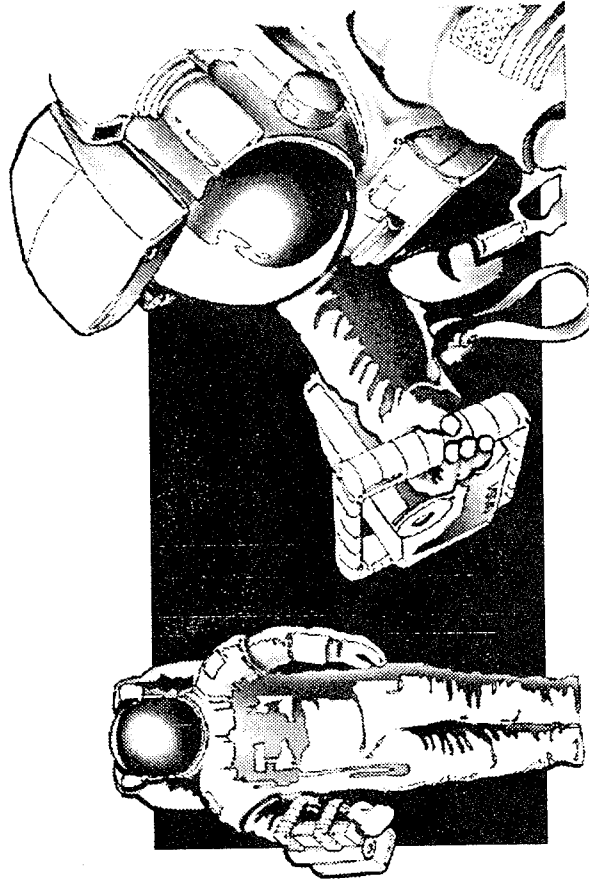
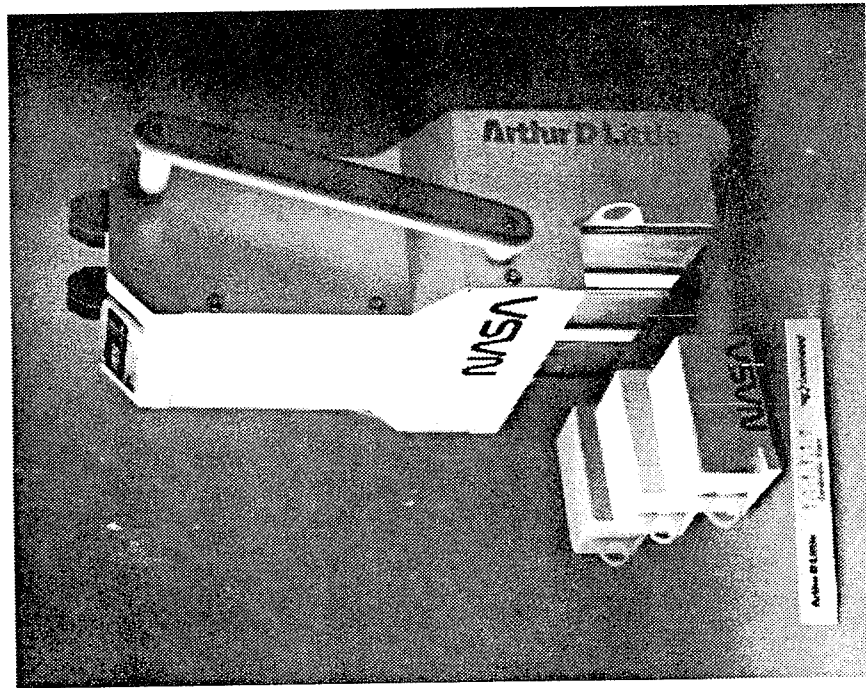
**Example: Soldier
Integrated Protective
Ensemble (SIPE)**



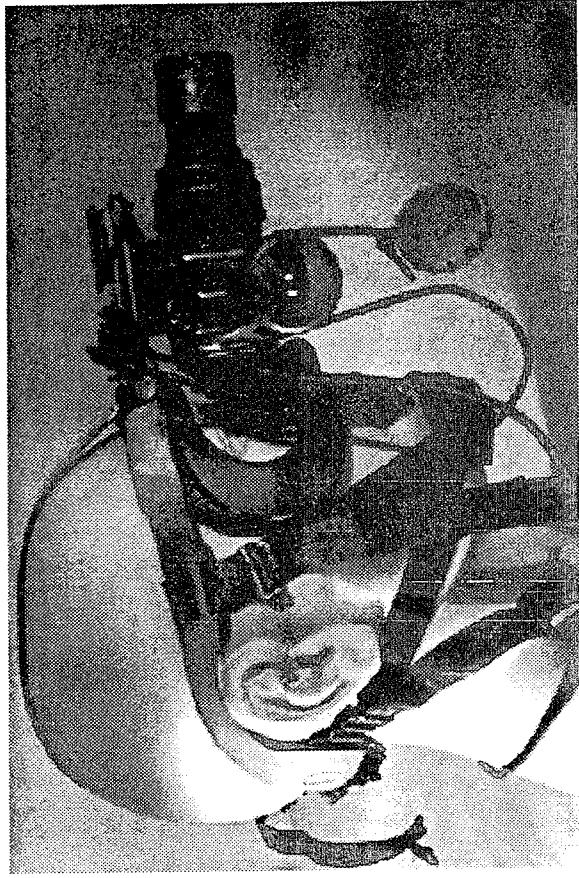
Example: 21 CLW—Generation II



Example: Hand-held Mass Spectrometer



Example: Night Vision System



System Integrator: Best to be a Generalist

- ◆ **A good symphony conductor can play all the instruments.**
- ◆ **The designer of the controls must understand all components to be controlled well enough to size them, specify them, select their operating points, provide for their needs, and suggest modifications to their designers.**

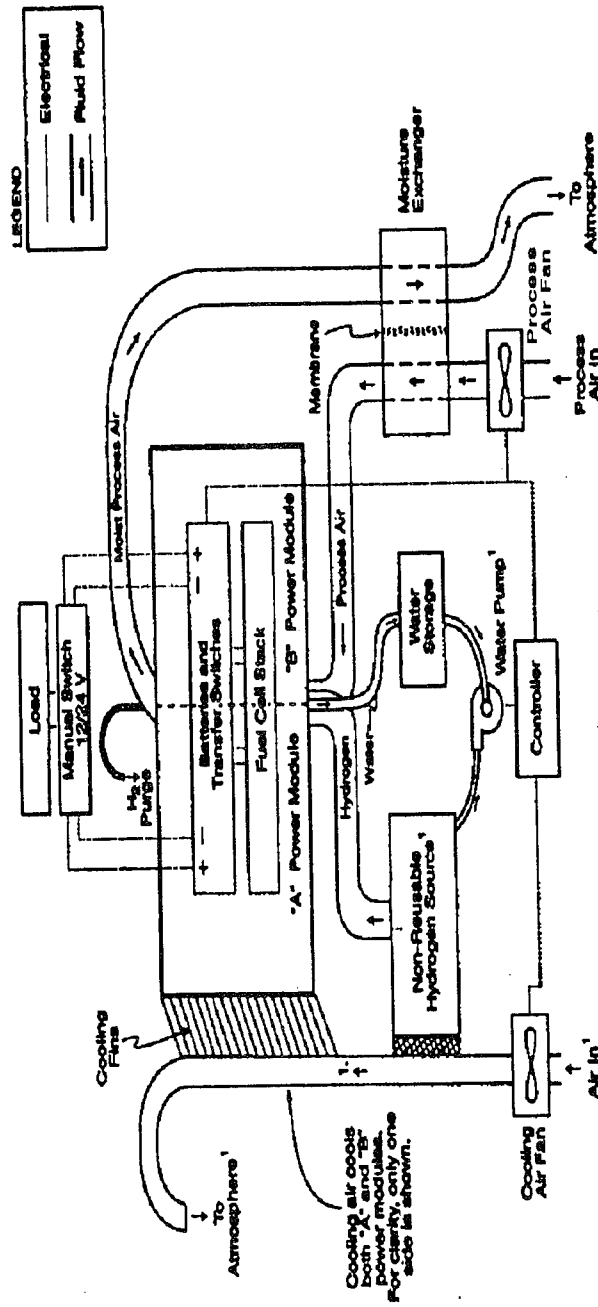
System Integrator must challenge specifications.

- ◆ **SI should define tradeoffs (e.g., quantify relationship between maximum ambient temperature and operating duration).**
- ◆ **SI should present these tradeoffs to the customer, thus giving the customer an informed choice.**

Portable fuel cell power systems present special challenges:

- ◆ **Obvious: weight, bulk, ruggedness, reliability, quietness**
- ◆ **Water management**
- ◆ **Thermal considerations**
- ◆ **Storage, transport, standby, start-up modes**
- ◆ **Safety**
- ◆ **Fuel gauge**

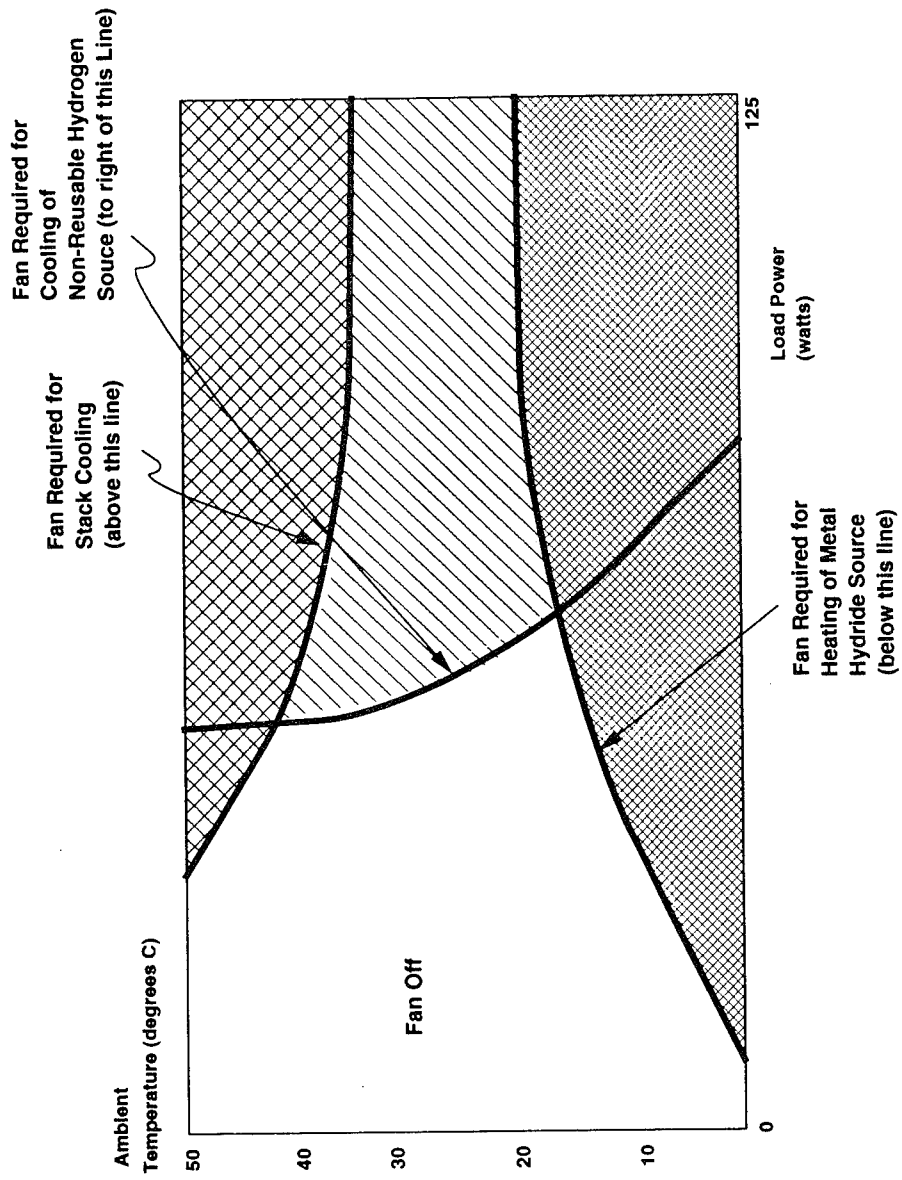
Fuel Cell Power Systems: More than a Fuel Cell Stack



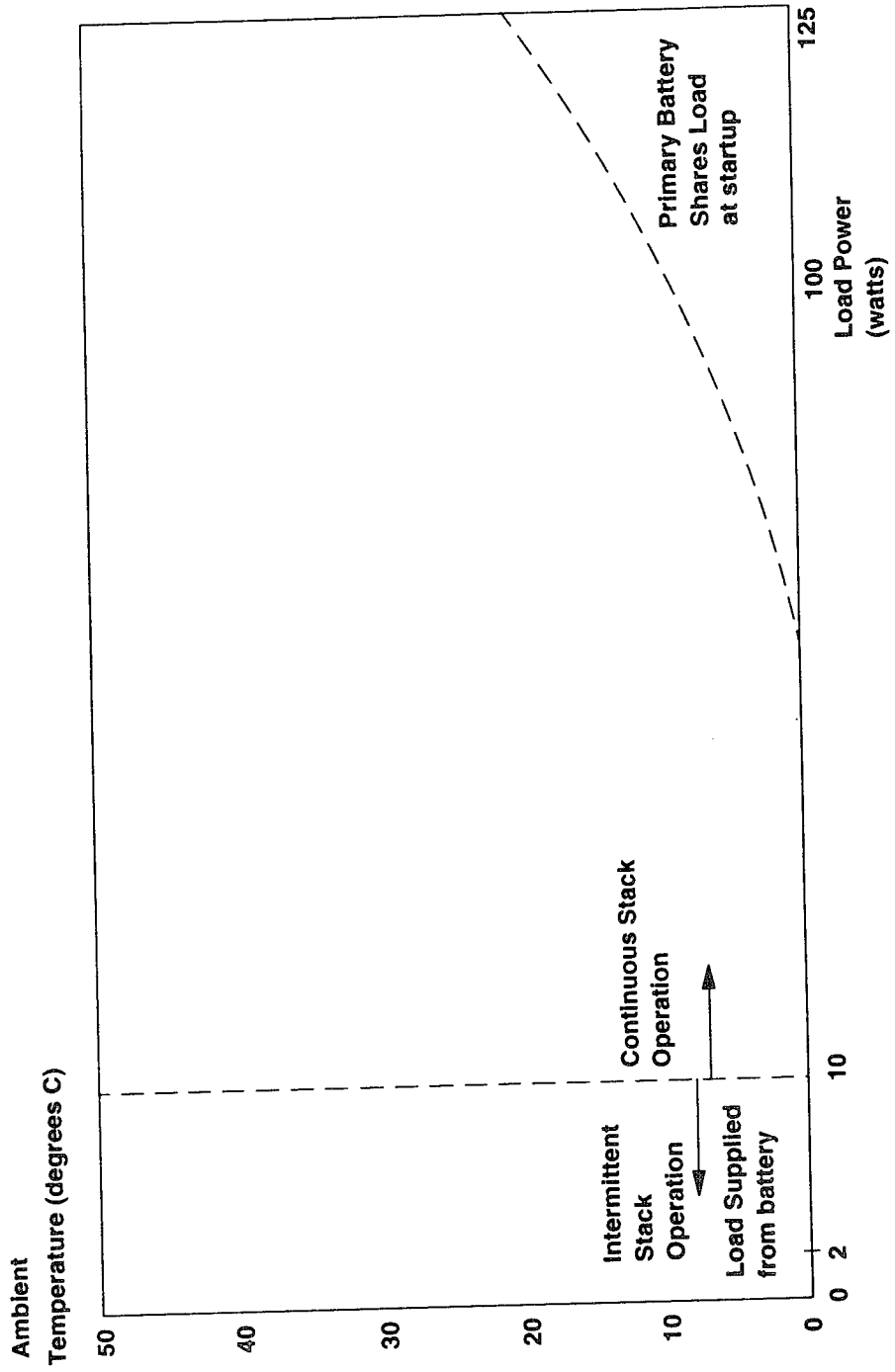
Example: Water Management

- ◆ **PEM fuel cell must be kept hydrated.**
- ◆ **Product water must not compromise secrecy of installation.**
- ◆ **Chemical hydride hydrogen supplies must be provided with water, at high pressure, in a controlled manner.**
- ◆ **Indigenous water may/may not be available/applicable.**

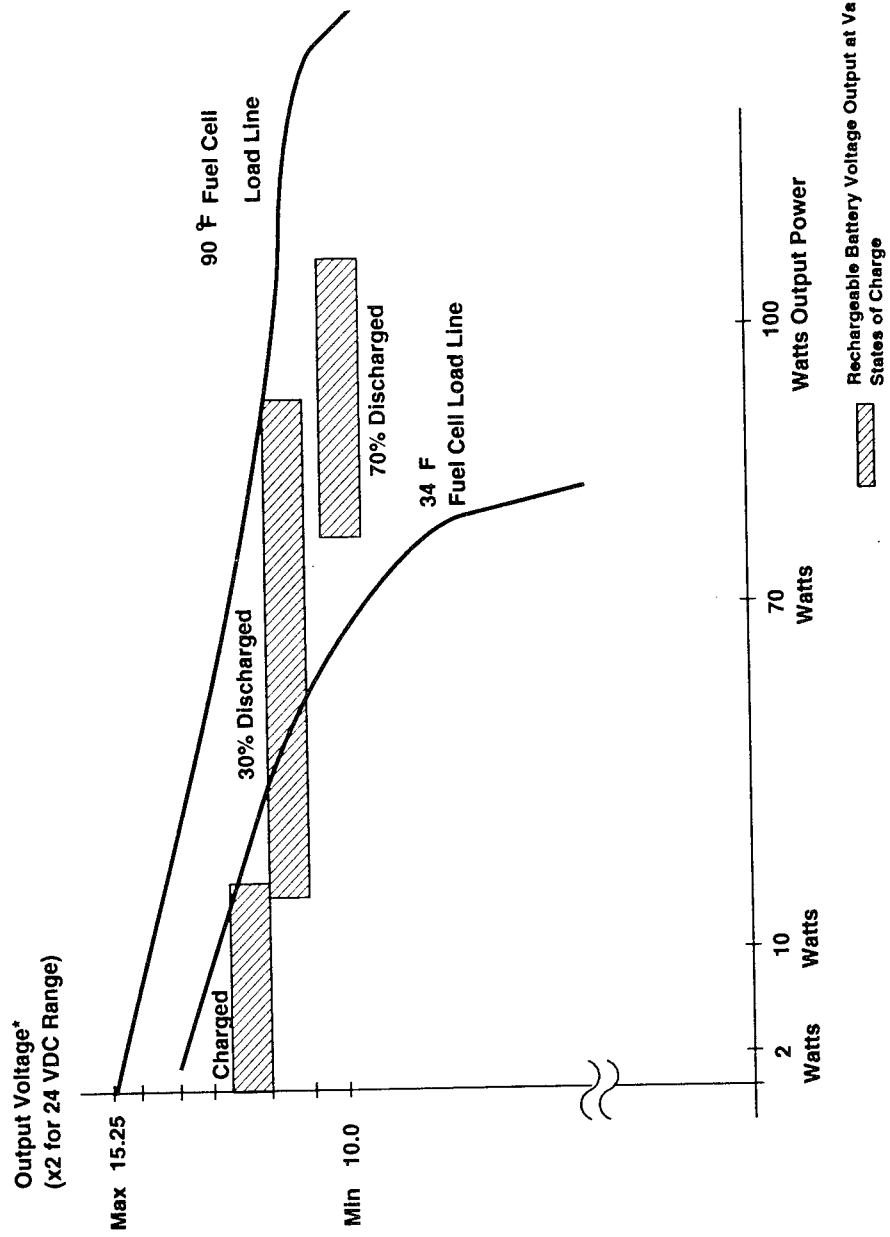
Example: Thermal Management



Example: Fuel Cell Stack vs Battery



Example: Balancing Charging and Discharging



ENERGY STORAGE: MANPACK HYDROGEN TANKS

Ira F. Kuhn, Jr.
Directed Technologies, Inc
4001 N. Fairfax Drive, Suite 775
Arlington, VA 22203

The author, in collaboration with two additional authors, Brian D. James and George N. Baum, conducted a study and submitted a report to the U. S. Army Research Office (ARO) on this particular subject. That unclassified report, a Phase I Final Technical Report, entitled "High Pressure Hydrogen Supply for Increased Fuel Cell Efficiency," was completed for ARO on September 30, 1993. The report's abstract is included as an added reference for the reader.

Phase I Final Report Abstract: "Fuel cells are being considered by the Army for future individual soldier power supplies. These power units are to be lightweight, compact and suitable for use on the battlefield. A compact, lightweight, safe source of pure hydrogen (and possibly pure oxygen) is urgently needed. The report examines a series of hydrogen/oxidant storage systems GH₂/Air (0 psig and 15 psig fuel cell operation), GH₂/GO₂, LH₂/Air (0 psig and 15 psig fuel cell operation), and LH₂/LOX. Compressed gas storage pressure is varied from 3,000 psi to 15,000 psi. Systems holding sufficient H₂/oxidant for 2.4 kW-hrs of fuel cell power are examined. An air compressive system operated by the expansion of high pressure gases is conceptually designed to allow "supercharging" of the fuel cell stack. System gravimetric and volumetric energy density is calculated for all system architectures."

A detailed technical report on various gas compression systems has been completed in recent years, and the use of compressed hydrogen gas was one of the proposed options in that report. The author reviewed the rationale and recommendations which were used to reach the conclusion that compressed hydrogen gas was a viable option for manpack power systems for the individual soldier. Specific tank storage requirements were discussed, and a chart displayed, which showed the relevance of producing a power system which could deliver 5 kW-hrs of energy storage from a pressurized manpack hydrogen tank. The tank's designed energy storage data was based upon the assumption of a Hydrogen/Air fuel cell burn rate of 0.124 lb H₂/kW-hr (.62 lb of H₂ for 5kW-hrs). A chart depicting the pressure-density relationship for compressed hydrogen was briefly explained to support the parameters used in the tank design. The author referred the Workshop participants to the full report for any additional details of interest in this manpack power supply concept.

Prospector VII

Small, Portable Fuel Cells for Military Applications

October 31 - November 3, 1994

Energy Storage: Manpack Hydrogen Tanks

by

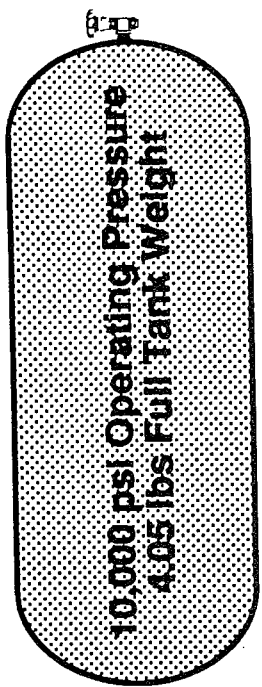
Ira Kuhn

Directed Technologies, Inc



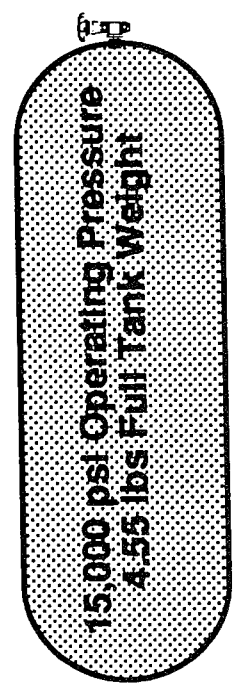
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TECHNOLOGIES, INC.

MANPACK HYDROGEN TANKS FOR 5 KW-HRS ENERGY STORAGE



6.25"

16.7"



5.4"

16.7"

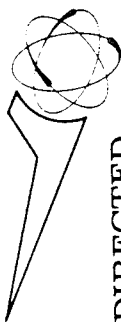
Volume	446 cu. in., 0.258 cu. ft.
Design Pressure	10,000 psi
Safety Factor	1.5
Burst Pressure	15,000 psi
Tank Weight	3.43 lbs
Figure of Merit	1.95 X 10 ⁶ inches
Hydrogen Weight	0.62 lbs
% H ₂ by Weight	15.3%
Hydrogen Density	2.40 lbs/ft ³

Volume	341 cu. in., 0.197 cu. ft.
Design Pressure	15,000 psi
Safety Factor	1.5
Burst Pressure	22,500 psi
Tank Weight	3.93 lbs
Figure of Merit	1.95 X 10 ⁶ inches
Hydrogen Weight	0.62 lbs
% H ₂ by Weight	13.6%
Hydrogen Density	3.15 lbs/ft ³

+ 1/2"

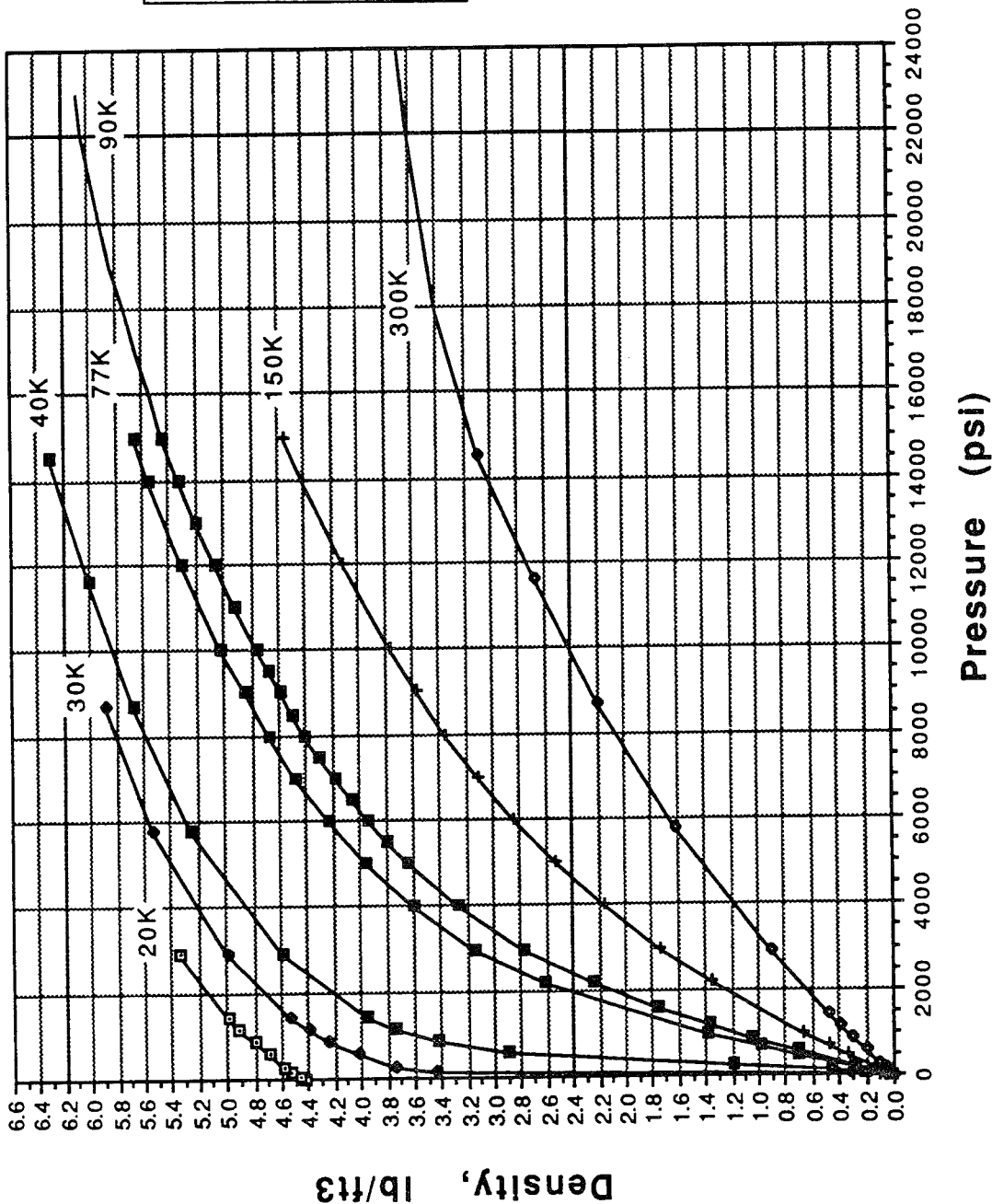
Tank dimensions are interior measurements.

* Energy based on H₂/Air fuel cell burn rate of 0.124 lb H₂/kW-hr.



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COMPRESSED HYDROGEN DENSITY



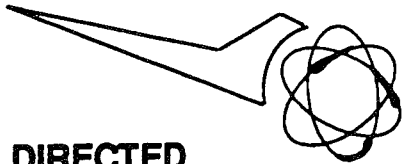
Densities of Interest

Pressure (psi)	Density (lbs/ft ³)	Temperature (°K)	Temperature (°C)
6700	4.42	77	-196
5000	3.96	77	-196
5000	3.66	90	-183
5000	2.95	120	-153
5000	2.53	150	-123
5000	2.01	200	-73
5000	1.67	250	-23
5000	1.43	300	27
3000	0.93	300	27

Typical Reformate Composition from Natural Gas

Contaminant Gas	% Vol.	Critical Point	
		Pressure (psia)	Temperature (°K) (°C)
H ₂	77.5	191	33 (-240)
CO ₂	18.8	1070	304 (31)
CH ₄	2.7	673	191 (-82)
∞	1	522	133 (-140)

Densities above 15,000 psi are estimated.



**DIRECTED
TECHNOLOGIES, INC.**

**HIGH PRESSURE HYDROGEN SUPPLY
FOR INCREASED FUEL CELL EFFICIENCY**

Revised

8 October 1993

**Prepared For:
Army Research Office (ARO)
P.O. Box 12211
Research Triangle Park, NC 27709-2211**

Contract No. DAAH04-93-C-0005

**Prepared By:
Directed Technologies, Inc.
4001 N. Fairfax Dr., Suite 775
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703/243-3383**

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1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE 30 September 1993	3. REPORT TYPE AND DATES COVERED Phase I Final Technical Report	
4. TITLE AND SUBTITLE High Pressure Hydrogen Supply for Increased Fuel Cell Efficiency		5. FUNDING NUMBERS	
6. AUTHOR(S) Brian D. James, George N. Baum, Ira F. Kuhn, Jr.			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Directed Technologies, Inc. 4001 N. Fairfax Dr., Suite 775 Arlington, VA 22203		8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P. O. Box 12211 Research Triangle Park, NC 27709-2211		10. SPONSORING/MONITORING AGENCY REPORT NUMBER	
11. SUPPLEMENTARY NOTES The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.			
12a. DISTRIBUTION/AVAILABILITY STATEMENT		12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Fuel cells are being considered by the Army for future individual soldier power supplies. These power units are to be lightweight, compact and suitable for use on the battlefield. A compact, lightweight, safe source of pure hydrogen (and possibly pure oxygen) is urgently needed. The report examines a series of hydrogen/oxidant storage systems GH ₂ /Air (0 psig and 15 psig fuel cell operation), GH ₂ /GO ₂ , LH ₂ /Air (0 psig and 15 psig fuel cell operation), and LH ₂ /LOX. Compressed gas storage pressure is varied from 3,000 psi to 15,000 psi. Systems holding sufficient H ₂ /oxidant for 2.4-10 kW-hrs of fuel cell power are examined. An air compression system operated by the expansion of high pressure gases is conceptually designed to allow "supercharging" of the fuel cell stack. System gravimetric and volumetric energy density is calculated for all system architectures.			
14. SUBJECT TERMS High Energy Density Fuels; Hydrogen Storage; Battlefield Power; The Ensemble Integrated Soldier System (TEISS) Power Unit			15. NUMBER OF PAGES 93
			16. PRICE CODE
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL

OXYGEN STORAGE

Jerry Chodil
Ball Aerospace, Cryogenics
Boulder, CO

The purpose of this briefing was to provide a qualitative system level comparison of several oxygen storage techniques that might be applied to fuel cell power system. For all but air breathing fuel cells, oxygen must be provided via some sort of storage system. The second chart (Oxidizer System Parameters) shows parameters that must be considered in making the choice of what oxygen source to select as a function of five different conventional methods of providing oxygen. This chart was constructed assuming that roughly 100 lbs of usable oxygen must be provided. Other usable oxygen quantities will provide different conclusions. The chart shows that one must consider the weight and volume of all the hardware associated with storing the oxygen before making a selection. As can be seen by "Weight Efficiency" and "Volumetric Efficiency" (defined in Chart #3), oxygen sources that have a high "Percent O₂ Liberated", don't necessarily come out favorably when the total system weight and volume are taken into account.

Chart #4 goes a step further to compare high-pressure oxygen-gas storage versus liquid-oxygen storage for fuel cell power systems that must provide various amounts of total energy. This chart shows specifically how much the total oxygen storage system weighs for various quantities of usable oxygen. The results are shown graphically in Chart #5. One can see that for total energy systems providing less than 60 KWH, high pressure gas (HPG) systems weigh less than liquid oxygen systems. In fact, at very low KWH, HPG systems weigh nearly an order of magnitude less. For systems providing more than 60 KWH, liquid oxygen storage systems weigh less than HPG systems.



Ball Corporation
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Systems Division

OXYGEN STORAGE

- High Pressure Gas
- Liquid

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Internet: jchodil@ball.com
11/1/94

Oxidizer System Parameters

Ball Corporation
Aerospace
Systems Division



Oxidizer System Parameters	High Pressure Oxygen (7000 psia)	Hydrogen Peroxide (H ₂ O ₂)	Potassium Superoxide (KO ₂)	Chlorate Candles (NaClO ₃)	Liquid Oxygen (LOX)
Density lb/ft ³	33.7	86.6	133.6	153	71.2
Percent O ₂ liberated	100	42	34	38	100
Lbs O ₂ /ft ³	33.7	36.4	45.4	58.1	71.2
Percent O ₂ with respect to LOX	47%	51%	64%	81%	100%
Weight Efficiency	.35 - .45	.3 - .4	.2 - .3	.2 - .3	.6 - .7
Volumetric Efficiency	.3 - .4	.4 - .5	.3 - .4	.4 - .5	.7 - .8

265



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DEFINITIONS

- **Weight efficiency:** Ratio of weight of stored oxygen to total oxidizer system weight

$$\frac{\text{(Stored oxygen mass)}}{\text{(Oxidizer system mass)}}$$

- **Volumetric efficiency:** Ratio of the volume of oxygen stored to the total oxidizer system volume.

$$\frac{\text{(stored oxygen volume)}}{\text{(system volume)}}$$



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Oxygen Storage System

Energy Requirement (kWhr)	Storage System					
	Oxygen	Hydrogen	High Pressure Oxygen (2)		Liquid Oxygen	
	Mass (1) (lb)	Mass (1) (lb)	Mass (lb)	Volume (cu ft)	Mass (lb)	Volume (cu ft)
1	0.8	0.1	1.4	0.046	22	0.081
3	2.4	0.3	4.2	0.14	25	0.16
10	8	1	14	0.46	32	0.34
30	24	3	42	1.4	53	0.78
100	80	10	139	4.6	122	2.1
300	240	30	411	14	308	5.4
1000	830	100	1361	46	947	16

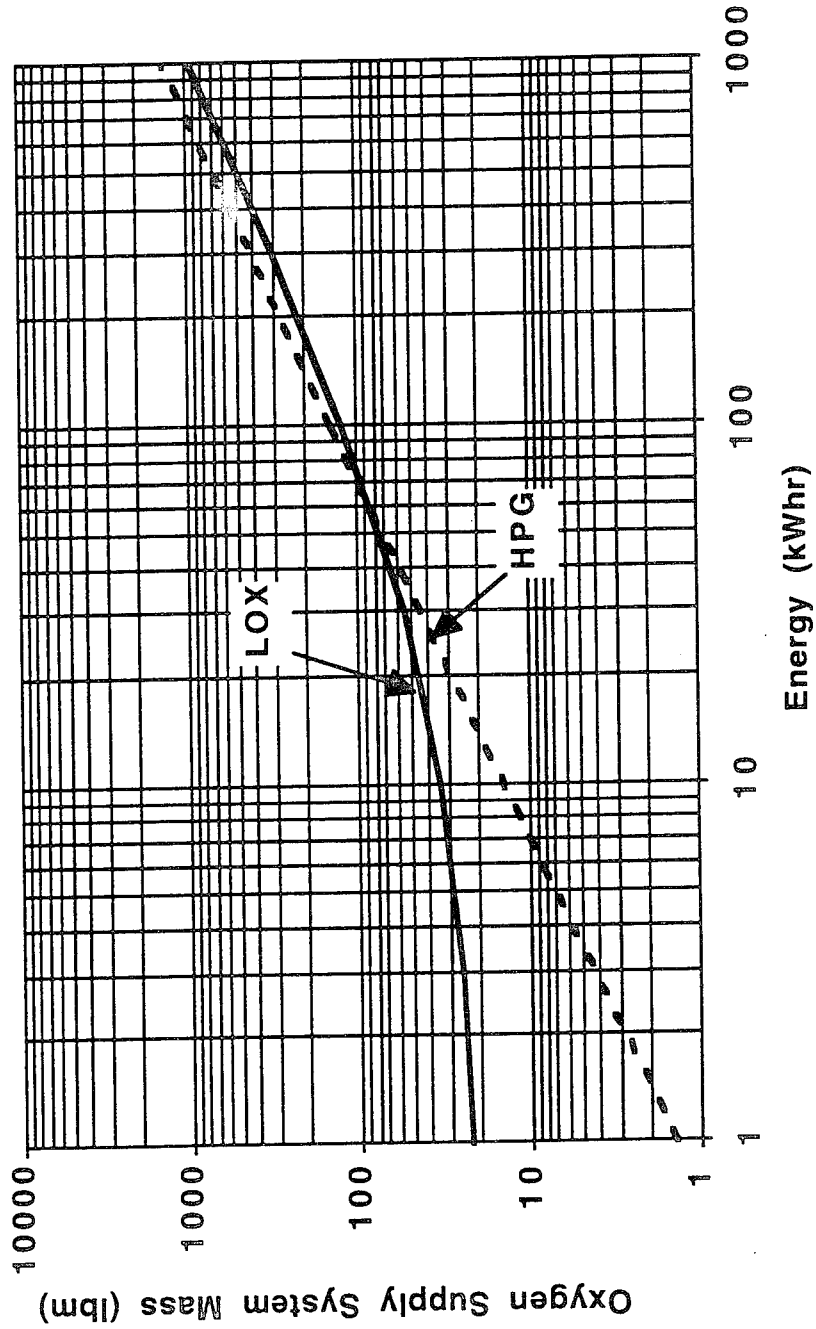
(1) Assumes 60% efficient fuel cell

(2) 3000 psi



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Oxygen Storage System



DIRECT METHANOL FUEL CELLS

Prepared for:
"Small Fuel Cells for Portable Power"
Workshop presented by
Army Research Office and Auburn University's Space Power Institute

October 1994

Robert F. Savinell
Department of Chemical Engineering and
The Ernest B. Yeager Center for Electrochemical Sciences
Case Western Reserve University
Cleveland, Ohio 44106

This discussion will be limited to the acid-type fuel cell based on using a proton exchange membrane. The fuel reaction or the anode reaction in this case combines methanol and water to produce carbon dioxide. The cathode oxygen, either from pure oxygen or air, is combined with protons to produce water. The overall reaction is a net water producer and its equilibrium voltage is around 1.186 v, therefore, somewhat less than the 1.23 v of a hydrogen oxygen cell. The major potential advantage with the direct methanol fuel cell is its energy density. As compared to the gaseous hydrogen oxygen fuel cell, its watt-hours per liter is ~ 3 orders of magnitude higher. Of course the energy density advantage would be somewhat less when hydrogen is in other forms such as liquid or solid. The direct methanol PEM fuel cell (DMPEMFC) has numerous advantages because of its high energy density, and because it can be readily stored and easily transported. The polymer electrolyte avoids electrolyte redistribution, causes no electrode flooding, enhances catalyst stability because of limited electrolyte content, and minimizes electrolyte-caused corrosion. A direct methanol fuel cell eliminates the need for a reformer. Therefore, the resulting power source is much lower in weight, smaller in volume, and is expected to have reduced costs. These systems will have a number of applications ranging from milliwatts to hundreds of kilowatts. At the low end of the power spectrum, applications include electronics and telephones. At the upper end of the spectrum, applications include automobiles, busses, trucks, trains, etc. In between there are many applications for both military to commercial uses.

During the past four to five years, there have been at least two workshops that dealt with the research issues which need to be addressed for developing a direct methanol fuel cell. The first meeting was sponsored by the Department of Energy and was held at the University of Georgetown in May 1990. The second more recent meeting was co-sponsored by DOE and ARPA and was held in Baltimore in April 1994. There are a number of recommendations coming from both of these workshops and most were very similar. In the more recent meeting though, there has been a recognition that further detailed studies are needed for understanding why a platinum-ruthenium catalyst system works well for the methanol oxidation reaction. Also, the

problem of methanol cross-over is much better understood and recognized to be a more important problem. In addition, the recent workshop emphasizes the need for developing oxygen reduction catalyst that are methanol tolerant. Also brought out was the fact that there is a need for reliable modeling of direct methanol fuel cell stack performance. Much of the advances in DMFCs in the past few years have come about because of an ARPA sponsored program. The state-of-the-art performance achieved by several contractors to ARPA are summarized in the transparencies. A .41v at 640 mA/cm² performance operating on a liquid feed 258 cm² fuel cell can produce 67 w per cell. Efficiencies have exceeded 25-30% at current densities less 300 mA/cm² and fuel utilization is greater than 70%. This program has now progressed beyond single cells and small multi-cell stacks are starting to be tested. As part of this program, IFC did a weight estimate for a small 250 watt DMPEMFC. They estimated a weight of about 10 lb. for this system. It is interesting that 40% of this weight comes from repeat parts. Presumably much of this weight is due to bi-polar plates and flow field components. Although there has been a number of successes in the ARPA program, there are still a number of challenges to be met in developing a direct methanol fuel cell.

The first challenge is poor anode kinetics. The anode reaction is slow because CO formed during the reaction strongly absorbs onto platinum surfaces. Of course, one can raise the temperature of the anode to reduce CO absorption. This is the approach being taken at Case Western Reserve University which will be discussed later. In addition, favorable catalyst can reduce the anode potential considerably. The best catalyst to date has been the platinum-ruthenium system which has a several hundred mV advantage over platinum, even up to a couple hundred mA/cm². Recent advances in developing this catalyst system has focused more on enhancing the surface area and utilization of the catalyst. Further improvements are still needed in enhancing the kinetics. Especially, the open circuit voltage needs to be reduce considerably for the anode reaction. Even with the best catalyst today, it is still about 200 mV positive of the reversible potential for methanol oxidation.

Another challenge for the development of the direct methanol fuel cell is the reduction of methanol cross-over. Methanol is highly soluble in water. Methanol permeates readily from the anode to the cathode where it reacts directly with oxygen. This cross-over results in the loss of fuel but also results in additional polarization at the cathode due to the mixed potential effect (~100 mV). There have been a number of strategies suggested to reduce this problem of methanol cross-over. Of course one strategy is to develop catalysts for oxygen reduction which are methanol tolerant. In another strategy, one tries to reduce the methanol solubility by using new electrolytes or lowering methanol activity. In the work at CWRU, higher temperatures are being considered which in effect lowers the activity of methanol and reduces cross-over from a level of ~100-300 mA/cm² down to 50 mA (Nafion phosphoric acid fuel cell at 150°C) and 10 mA/cm² for new electrolytes (PBI).

Some groups are taking other approaches to reducing methanol cross-over. One approach is to use thick electrodes; the idea is to consume as much methanol in the electrodes so there is little left to cross-over. This is feasible except when one does this, the catalyst utilization is reduced substantially. Another approach is to include barrier layers or to deposit pore blockers into the membrane pores. The idea here is to block some of the pores with an inert material so that

methanol diffusion is restricted. Of course, the conductivity of the polymer is directly related to the water content in the pores of the Nafion. Therefore, a reduction in permeability usually will correspond to a reduction in conductivity. We have examined the trade-off by a simple cross-over model which shows that placing pore blockers into a Nafion membrane will have little beneficial effect except at low current densities.

At Case Western Reserve University, as I mentioned earlier, we have a program to develop direct methanol fuel cells. Our program is based on the idea of operating at higher temperatures where we can reduce the anode poisoning and also reduce methanol cross-over by lowering methanol activity in the polymer. This program has a number of objectives. A major objective is to develop proton exchange membranes which can operate at higher temperatures, namely the 150°-200°C range. In addition, we are developing new catalyst systems for both the methanol electrode and the oxygen electrode, in particular oxygen reduction catalyst which are methanol tolerant. We are pursuing fundamental studies to further understand the details of the kinetic mechanism of methanol oxidation.

One of the major accomplishments of this program has been the identification of a basic polymer known as polybenzimidazole (PBI) which can be doped with an acid to yield proton conductivity at temperatures up to 200°C. This polymer has another advantage in that it is a homogeneous phase material which tends to provide a good barrier against the permeation of methanol. In experiments done in our laboratory, we have shown that the equivalent current density of methanol cross-over can be reduced to $<10 \text{ mA/cm}^2$ with these materials. PBI fuel cells have been built and operated at Case Western Reserve University. Although we have not achieved levels of performance as the best results reported by other laboratories for the lower temperature system, our results are very encouraging because we have been able to operate at higher temperatures with very little extra humidity and operational problems. We have shown stable reproducible operation. With development of optimum electrode structures for this polymer, performance should increase substantially. We have recently submitted membranes to an industrial firm and results with 25 cm^2 cells have demonstrated very good performance considering their electrodes have not been optimized.

In summary, progress towards developing efficient and compact direct methanol fuel cells is very encouraging. Much of these advances have been made during the past several years in a program sponsored by ARPA. However, there are still advances needed in enhancing the performance of all fuel cell components. Also, greater understanding is needed of the fundamental processes. Engineering design must now be addressed, and a focus must be given to reducing costs.

PROSPECTOR VII WORKSHOP

“SMALL FUEL CELLS FOR PORTABLE POWER”

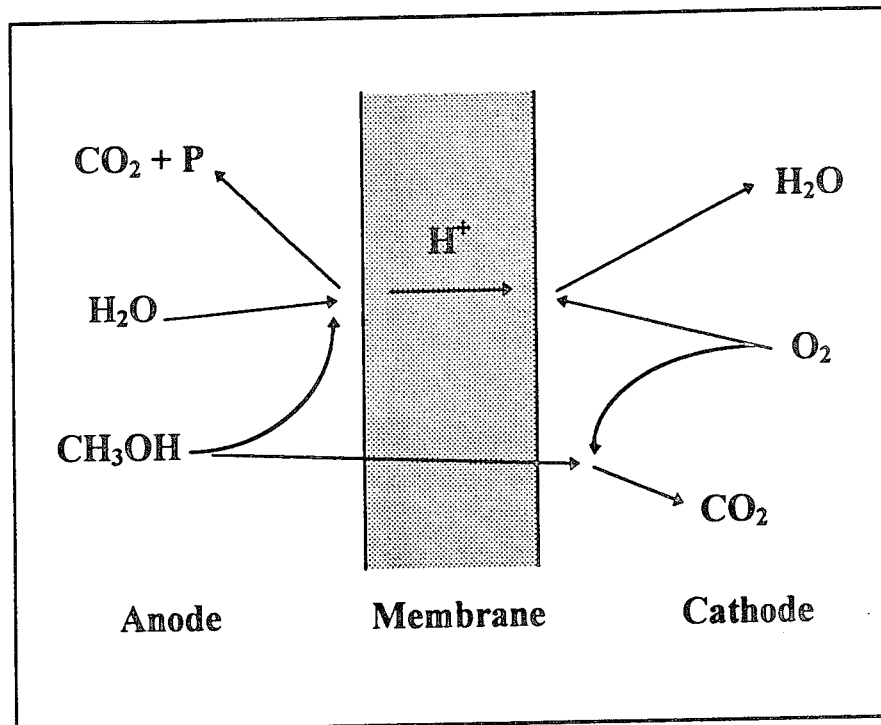
October 31- November 3, 1994

Durham, North Carolina

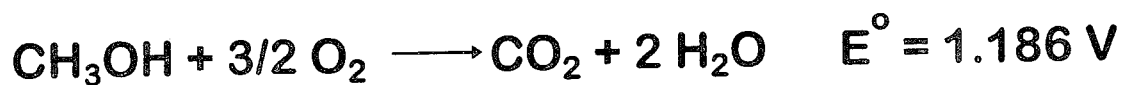
**Sponsored by:
Army Research Office and
Auburn University' Space Power Institute**

**Robert F. Savinell
Professor of Chemical Engineering and
Director, Ernest B. Yeager Center for
Electrochemical Sciences
Case Western Reserve University
Cleveland, Ohio 44106**

DIRECT METHANOL PROTON EXCHANGE MEMBRANE FUEL CELL (DMPEMFC)



OVERALL REACTION



COMPARISON OF ENERGY DENSITY

Couple	Wh / Kg	Wh / l (25°C, 1atm)
H_2 / O_2	39,700	3.5
$\text{CH}_3\text{OH} / \text{O}_2$	6,307	4,982



DMPMF

ADVANTAGES

FUEL

- HIGH ENERGY DENSITY
- EASY TO STORE
- EASY TO TRANSPORT

PEM

- AVOID ELECTROLYTE REDISTRIBUTION
- NO ELECTRODE FLOODING
- ENHANCE CATALYST STABILITY
- MINIMIZE ELECTROLYTE CAUSED CORROSION

ELIMINATES

- LOWERS WEIGHT

REFORMER

- LOWERS VOLUME
- LOWERS COST



DMPMEMFC

APPLICATIONS

mW → W → kW → 10 kW → 100 kW



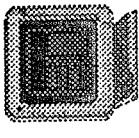
TELEPHONES



NAVIGATION BUOYS



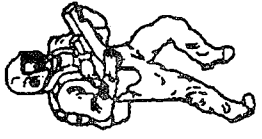
TOOLS



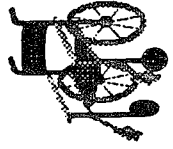
COMPUTERS



ELECTRONICS



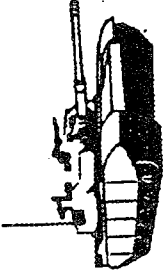
SOLDIERS



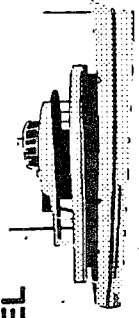
WHEELCHAIRS



RECREATIONAL VEHICLES



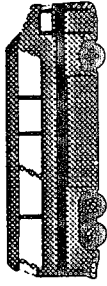
TANK HOTEL



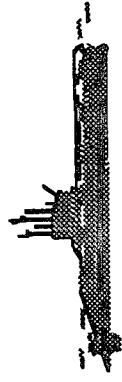
YACHTS



AUTOMOBILES



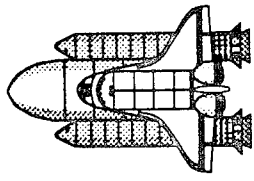
BUSES



UUV



GOLF CARTS



SPACE



DIRECT METHANOL FUEL CELLS

WORKSHOPS

DOE: Georgetown University
May 1990

DOE/ARPA: Baltimore, Maryland
April 1994

Recommended Areas of Research:

- Kinetic and Mechanistic Studies
- New Materials as Electrocatalysts for Methanol Oxidation
- Surface Sensitive Techniques for Methanol Electrocatalysts
- Electrolytes
- Systems Considerations
- Methanol Tolerant O₂ Catalysts*
- Reliable Modeling of DMFC Stack Performance*

*April Meeting

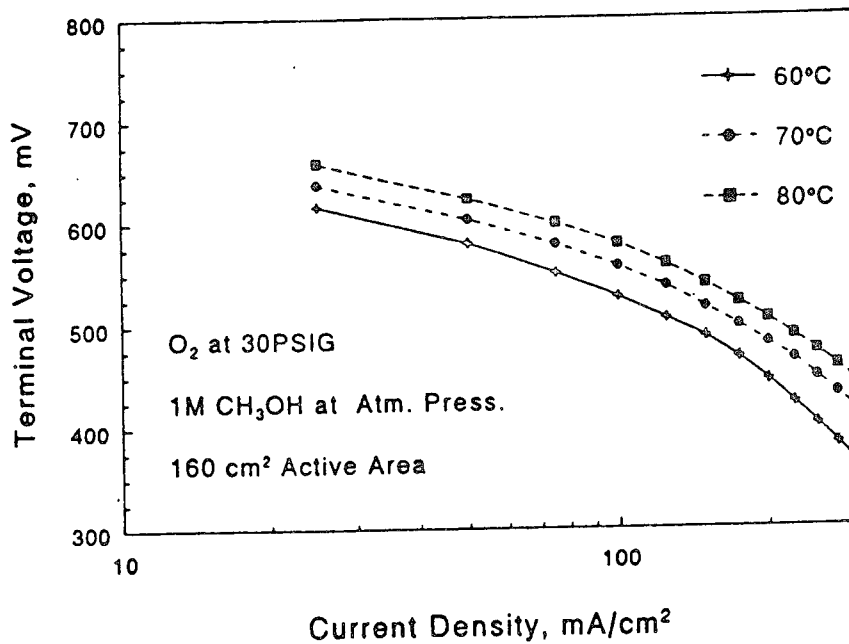


DMPEMFC

STATE-OF-THE-ART PERFORMANCE

ARPA DIRECT METHANOL FUEL CELL PROGRAM

<u>Single Cell:</u>	0.33 V @ 800 mA/cm ² 0.41 V @ 640 mA/cm ² 258 cm ² = 67 W/Cell	Life Test	<u>Single Cell:</u>	0.45 V @ 300 mA/cm ² for >200 hrs. continuous >500 hrs. intermittent
<u>4-Cell Stack:</u>	0.4 V @ 430 mA/cm ² 258 cm ² = 178 W		<u>5-Cell Stack:</u>	0.56 V @ 100 mA/cm ² (48 W) for > 400 hrs.
<u>Efficiency:</u>	25-30% @ <300 mA/lm ²			
<u>Fuel Utilization:</u>	>70% @ 300 mA/cm ²			



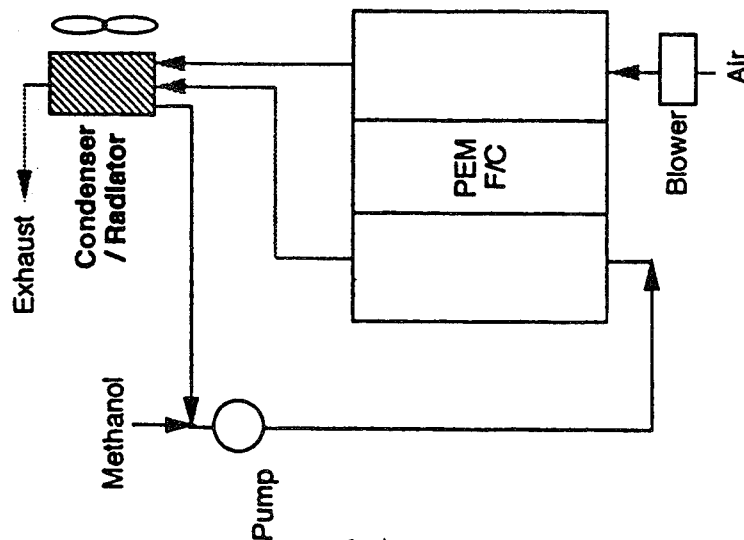
Effect of Temperature on Performance of 160 cm²
Direct Liquid Methanol PEMFC. Giner, Inc. 10/93.



IFC DMPEMFC WEIGHT ESTIMATE

250 watts , 1000 watt-hrs
0.6 v/c , 250 ASF

Net DC power	250 watts
Gross DC power	260 watts
LHV Efficiency (@ 0.6 v/c, 250 asf)	43.2 %
Stack voltage	12 volts
Condenser Heat Rejection	1019 btu/hr
MeOH Input	0.2306 lbs/hr
MeOH in exhaust	0.0023 lbs/hr
Assumed MeOH Crossover	50 asf equivalent
Air utilization	49.4 %
Cell temperature	170 °F
Cell Pressure	15 psia
Condenser Exhaust Temperature	123 °F



COMPONENTS	Weight (lbs)
Stack : Repeat Parts	4.22
Stack : Non-Repeat Parts	1.24
CH3OH for power	0.76
CH3OH crossover	0.15
CH3OH vented	0.01
CH3OH tank	0.05
Radiator	1.21
Radiator Fan	0.85
Process Air Fan	0.35
Methanol-H2O pump	0.94
Miscellaneous	0.49
TOTAL LBS	10.27

ARPA DMFC PROGRAM

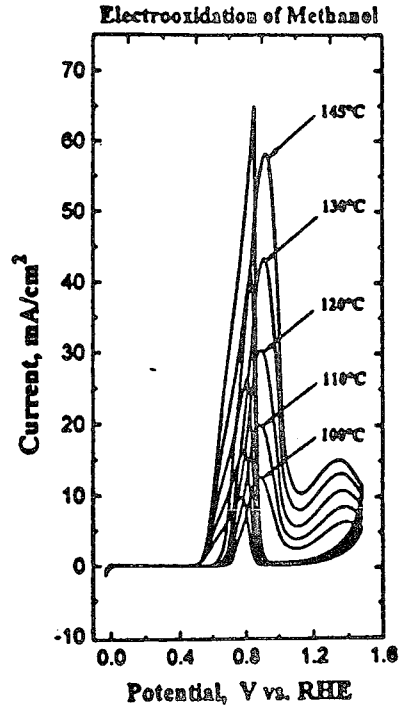
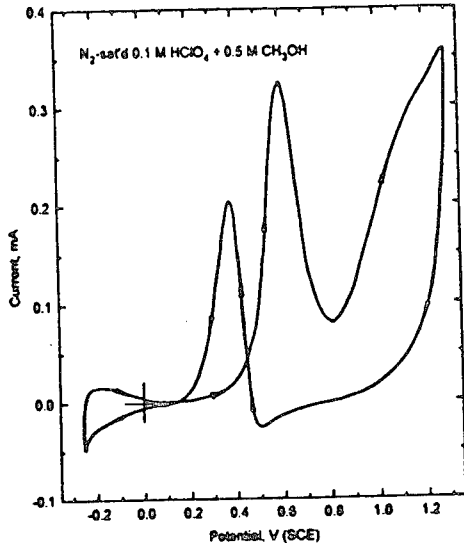
REVIEW 4/94



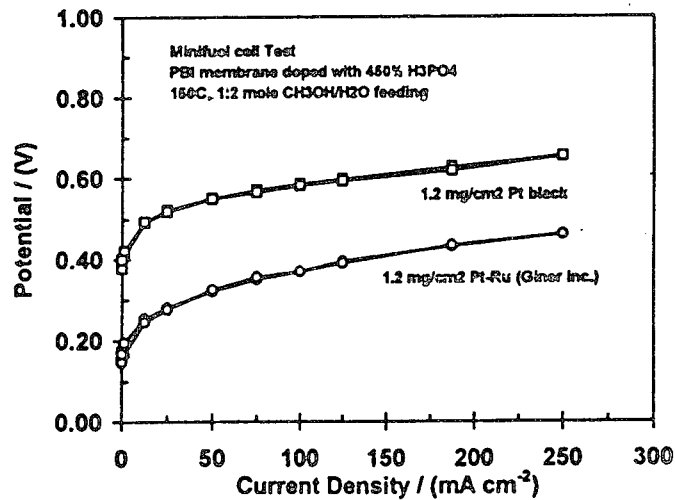
DMPEMFC

CHALLENGES

POOR ANODE KINETICS



STATUS: Pt/Ru Best Catalyst to Date



**NEEDS: Kinetics Need Further Improvement
Lower OCV**



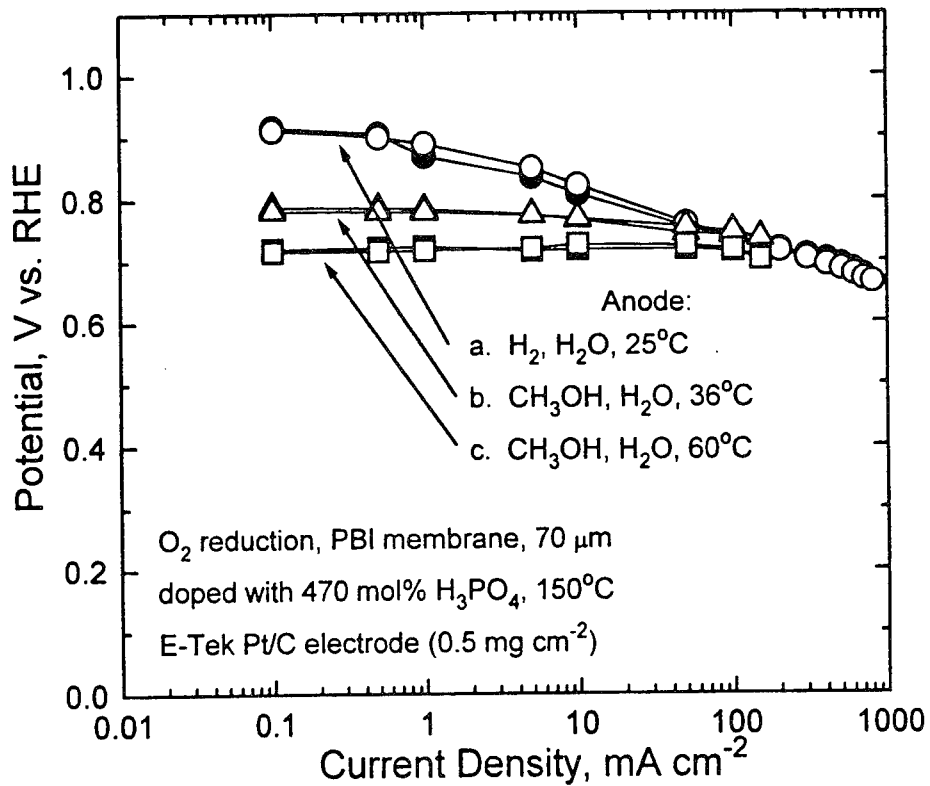
DMPEMFC

CHALLENGES

- METHANOL CROSSOVER
- FUEL LOSS

$$\text{Efficiency} = \frac{I_{\text{cell}}}{I_{\text{cell}} + I_{\text{crossover}}}$$

- CATHODE POLARIZATION

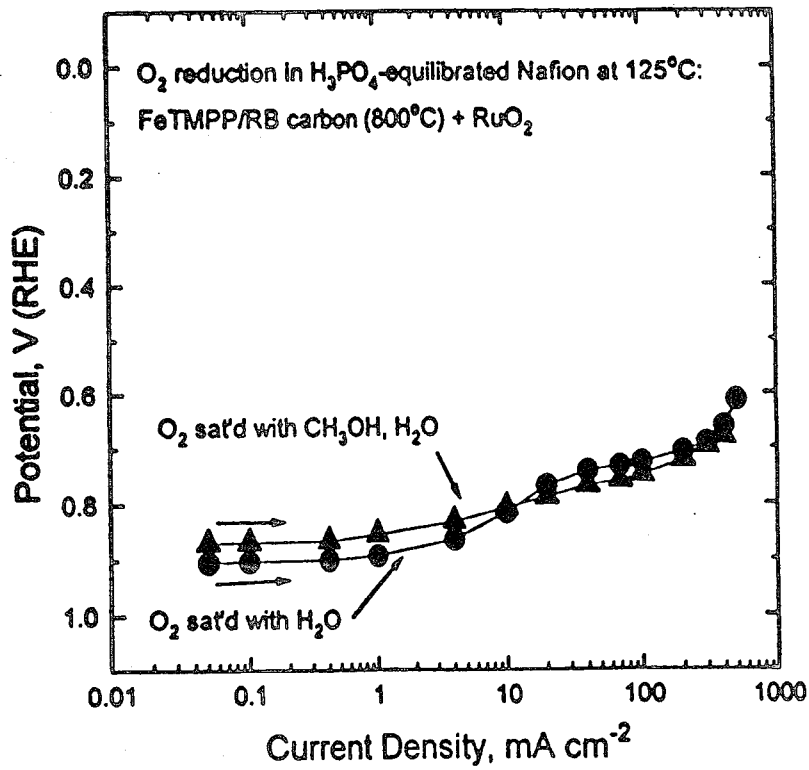


DMPEMFC

METHANOL CROSSOVER

STRATEGIES

- Methanol Tolerant Cathodes



DMPEMFC

METHANOL CROSSOVER

STRATEGIES

- Reduce Methanol Solubility

New Electrolytes

Lower methanol Activity

Membrane Electrolyte	Temperature °C	Methanol Crossover mA/cm ²
Nafion/H ₂ O	60	100 - 300
Nafion/H ₃ PO ₄	150	50 - 100
PBI/ H ₃ PO ₄	150	10

- Reduce Methanol Diffusion

Thick Electrodes

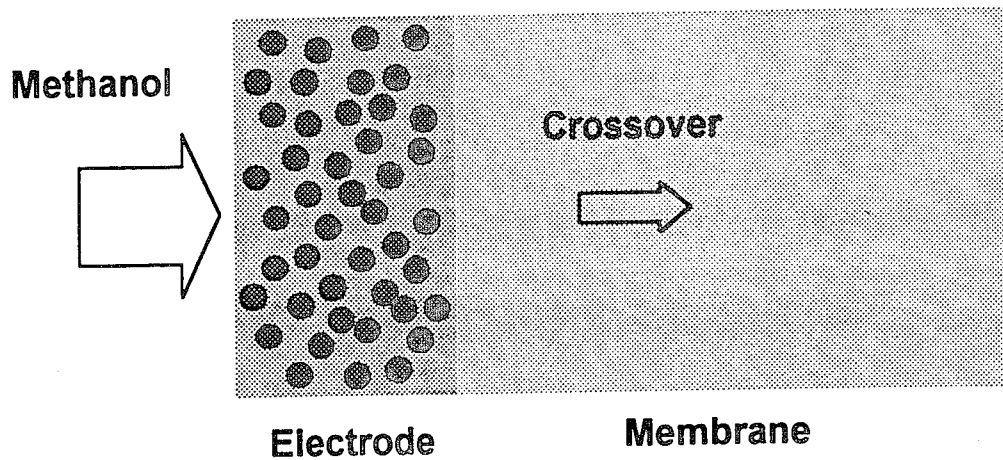
Barrier Layers:

Composite Materials

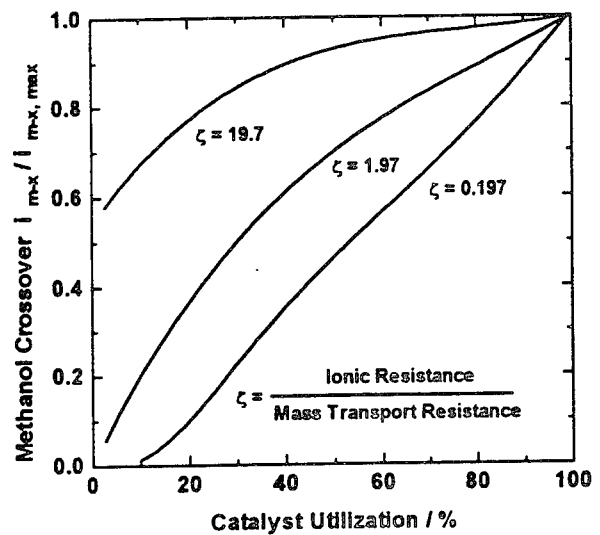
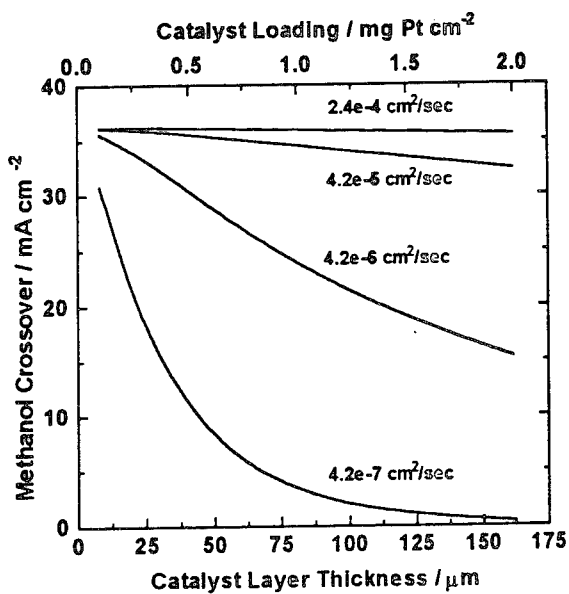
Pore blockers



REDUCE METHANOL CROSSOVER WITH THICK ELECTRODES



Thick Electrodes → Lower Crossover → Reduce Catalyst Utilization

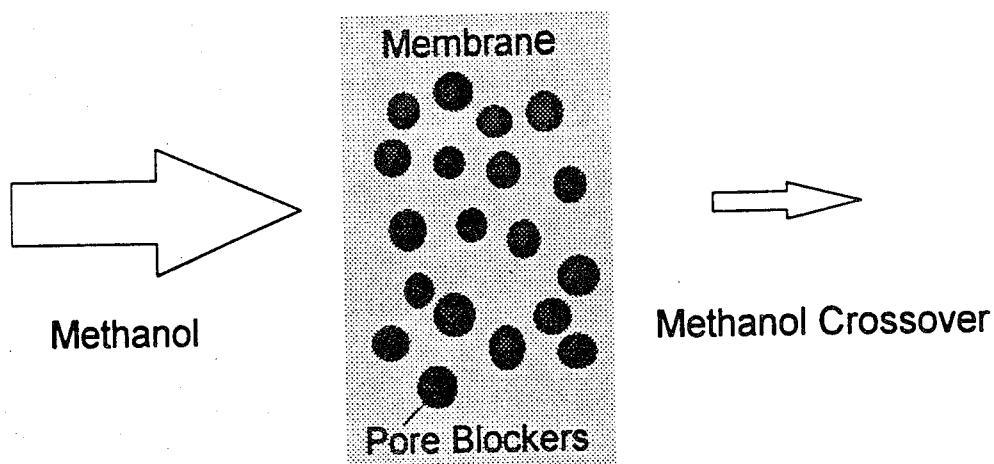


J-T Wang and R.F. Savinell, *Electrochemical Society Spring Meeting, San Francisco, May '94*



REDUCE METHANOL CROSSOVER WITH MODIFIED MEMBRANE

MODEL CALCULATION



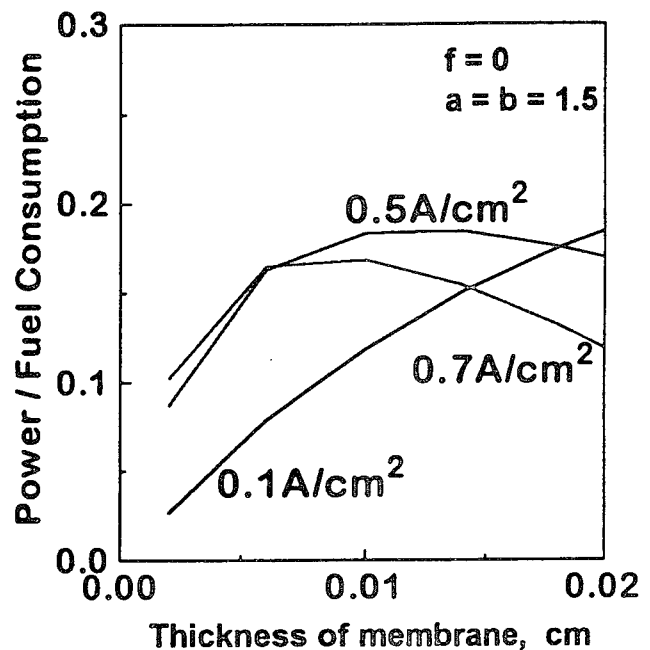
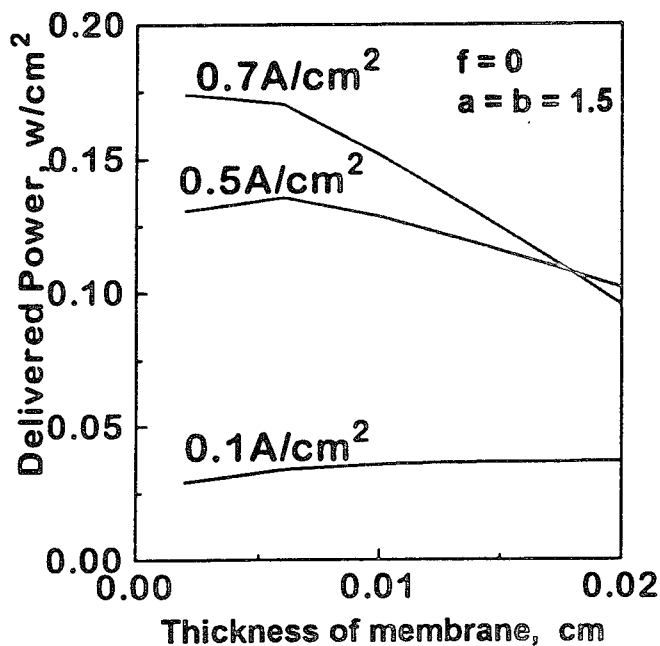
ASSUMPTIONS:

- Cathode kinetics
with mixed potential effect
- Membrane conductivity
- Methanol permeability
- Constant anode potential
- Based on Nafion at 60°C



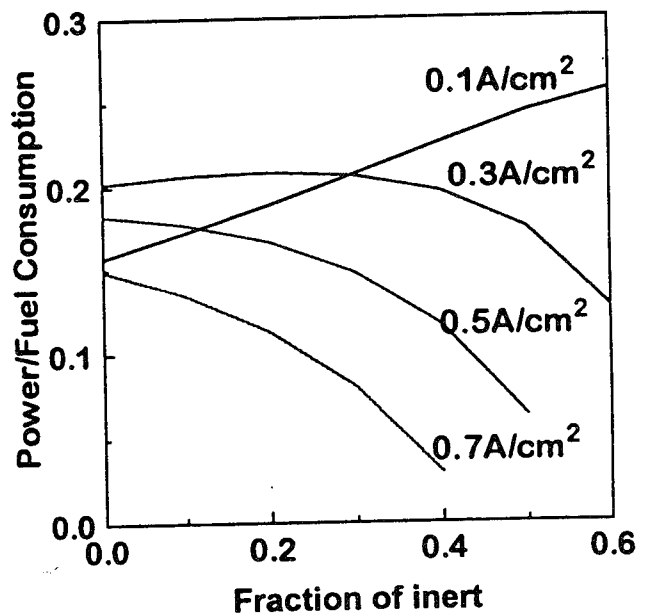
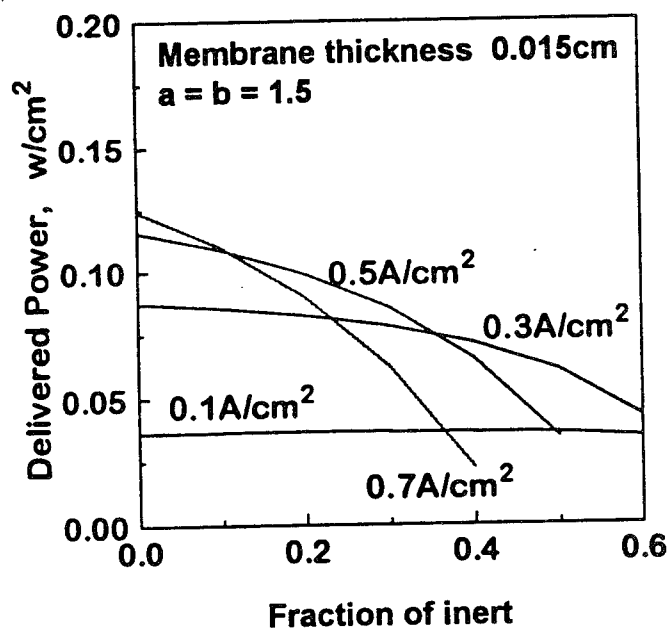
CROSSOVER VS FILM THICKNESS

- Low current density:
Increase fuel utilization,
No effect on power.
- Median current density:
Shallow optimum fuel utilization,
Decrease power.
- High current density:
Thinner optimum thickness.



CROSSOVER VS ADDED PORE BLOCKERS

- Ineffective except at low current density.
- Could be effective if pore blockers do not reduce conductivity.





PROGRAM OBJECTIVES

- Develop an efficient proton-exchange-membrane (PEM)-based methanol/air fuel cell capable of operating at 200°C to provide a compact and reliable energy source for military applications.
- Identify, synthesize and test new polymers, catalysts, and electrode structures to advance the fuel cell development program.
- Expand fundamental understanding of chemical and physical phenomena in fuel cells through theoretical and experimental studies leading to next generation advances in PEM fuel cell technology.

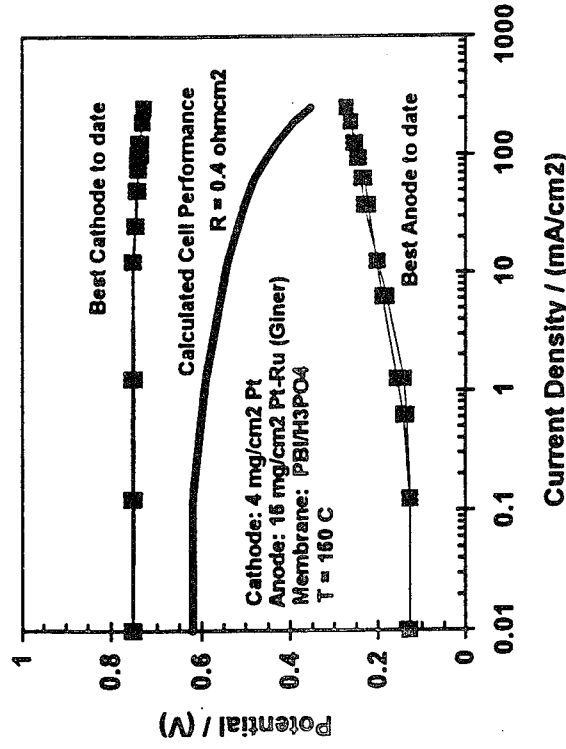
ACCOMPLISHMENTS

- Identified two proton conducting polymers capable of operating at 200°C under low humidity conditions.
- Synthesized a number of perfluorinated proton conducting polymers with properties nearly equivalent to present day state-of-the-art materials but with much more synthesis flexibility to alter physical and transport properties.
- Developed an oxygen reduction catalyst with activity equivalent to platinum and tolerant to the presence of methanol.
- Advanced experimental methods for model catalyst studies and achieved further theoretical understanding of methanol catalyst mechanism.

APPROACHES

- Synthesize, characterize and test electrochemical performance of novel ion conducting polymers.
- Perform theoretical and experimental studies to identify catalyst for methanol oxidation and oxygen reduction at 200°C in a polymer environment and elucidate kinetic mechanisms.
- Develop mathematical models, test and optimize electrode structures and fuel cell assemblies.

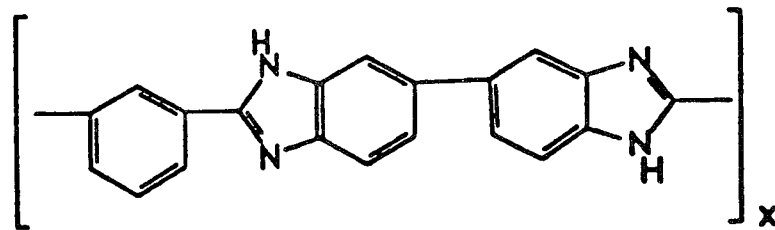
METHANOL/OXYGEN PBI FUEL CELL PERFORMANCE



- Further improvements in cell performance anticipated from improved cathode structure, lower cell resistance

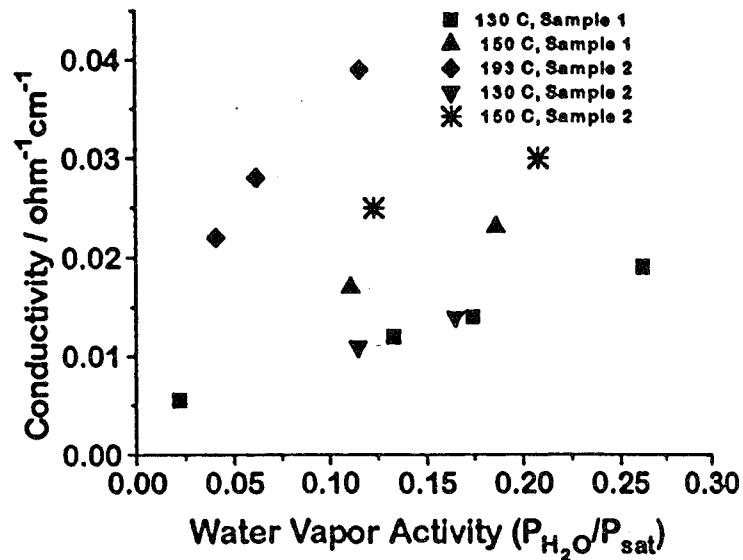
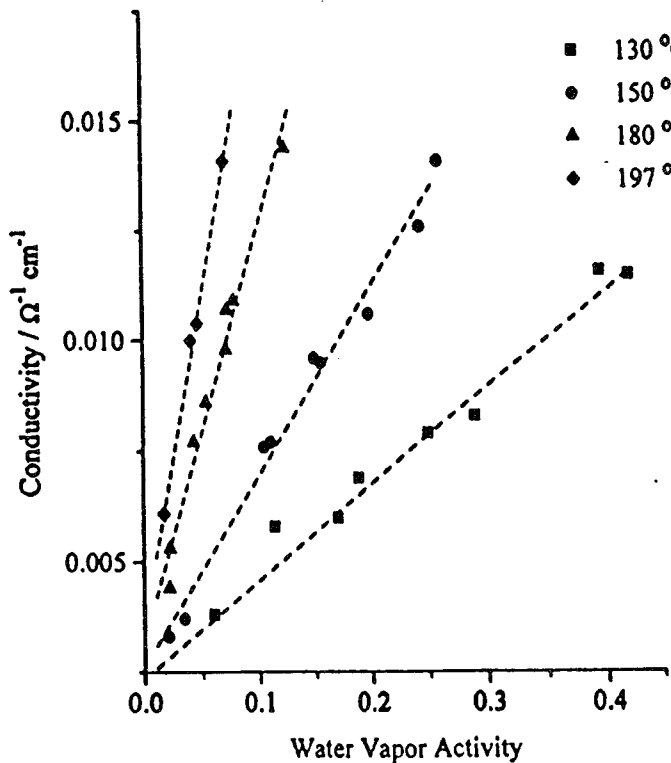
POLYBENZIMIDAZOLE

- Polybenzimidazole polymers have good thermal stability, high mechanical strength and excellent vapor barrier properties.
- Doping of polybenzimidazoles with phosphoric acid provides proton conductivity, improved thermal stability, greater flexibility.
- Preliminary results with polybenzimidazole membranes in H_2/O_2 fuel cells have shown high performance, low sensitivity to humidification conditions.



338 mole% H_3PO_4

501 mole% H_3PO_4



Permeability of doped polybenzimidazole

Doping	Gas	Temperature °C	Permeability	Equivalent CD / mA/cm ²
H ₂ SO ₄	Methanol	85	175, 320	7, 13
H ₂ SO ₄	Methanol	130	139, 156	5, 6
H ₃ PO ₄	Methanol	130	183, 186	7, 7
H ₃ PO ₄	Methanol	155	270	11
H ₂ SO ₄	Water	130	9000, 10000	-
H ₃ PO ₄	Water	140	4400	-
H ₃ PO ₄	Hydrogen	150	180	2
H ₂ SO ₄	Oxygen	125	11, 20	0.3, 0.6
H ₃ PO ₄	Oxygen	140	10	0.3

Permeability Coefficients are given in barrer

1 barrer = 10^{-10} cm³(STP) cm / cm² s cmHg

Multiple entries indicate results from multiple samples.

Equivalent current densities assume a one atmosphere pressure differential.

H₂SO₄ doping level 247 mol%

H₃PO₄ doping level 338 mol%

- Methanol permeability significantly lower than in Nafion
- Oxygen permeability similar to phosphoric acid



DMPEMFC

SUMMARY

- PROGRESS TOWARDS DEVELOPING EFFICIENT AND COMPACT DIRECT METHANOL FUEL CELL IS ENCOURAGING.
- ADVANCES STILL NEEDED IN PERFORMANCE OF ALL COMPONENTS, IN UNDERSTANDING FUNDAMENTAL PROCESSES , IN ENGINEERING DESIGN AND IN COST REDUCTION.



"Evaluation and Modelling of PEM Fuel Cells"

Summary of 1 Nov 94 Presentation to "Workshop on Small Fuel Cells for Portable Power" by Dr. Ronald F. Mann, Royal Military College of Canada.

An overview was presented of the power source-related work of the RMC Electrochemical Power Sources and Air Independent Propulsion Groups. Major activity areas are:

- PEM Fuel Cell
- Hydrogen Supply & Storage for Fuel Cells
- Oxygen Supply & Storage for Fuel Cells
- Non-PEM Fuel Cell Activities

It was emphasized that most of the work over 10 to 15 years has been focussed on kW and multi-kW power requirements.

The PEM work has concentrated on the evaluation of Ballard Power System hardware and the determination of performance limitation of Ballard stacks. An "electrochemical performance model" has been developed and applied over the range from small single cells to large multi-cell stacks. This model is essentially a steady state, constant temperature, relationship linking stack voltage, current, temperature, pressure, concentration of anode feed gas, concentration of cathode feed gas, and both anode and cathode gas stoichiometre ratios (% excesses) in the feed. The model implicitly assumes that the operating conditions are such that water management in the stack is under control.

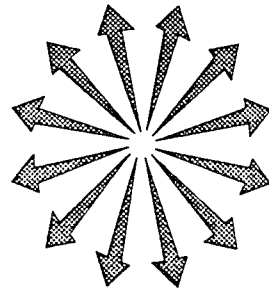
Currently a "steady state thermal model" of a PEM stack is being developed. This will be extended to a "dynamic thermal model" which will then be coupled to the "electrochemical performance model" to give an overall stack dynamic performance model.

At present, most of the work is funded by the Canadian Navy.

EVALUATION AND MODELLING OF PEM FUEL CELLS

Presentation to Workshop on Small Fuel Cells for Portable Power
Durham, North Carolina, October 31 - November 3, 1994

Dr. Ronald F. Mann



Electrochemical Power Sources Group

Royal Military College of Canada
Department of Chemistry and Chemical Engineering
Kingston, Ontario, CANADA K7K 5L0

Overview of Our Activities

- PEM Fuel Cell
- Hydrogen Supply & Storage for Fuel Cells
- Oxygen Supply & Storage for Fuel Cells
- Non-PEM Fuel Cell Activities
- Comments re Funding



FUEL CELL ACTIVITIES

- GENERAL MONITORING OF FUEL CELL DEVELOPMENT PROGRAMS IN THE WORLD WITH A MAJOR EMPHASIS ON PROTON EXCHANGE MEMBRANE (P.E.M.) FUEL CELLS (ie. descendants of earlier solid polymer electrolyte (SPE) fuel cells)

- MAJOR TECHNICAL INVOLVEMENT IN CANADIAN GOVERNMENT-FUNDED PROGRAMS AT BALLARD POWER SYSTEMS OF NORTH VANCOUVER

- RECEIVE BALLARD HARDWARE FOR EVALUATION AND FOR USE IN RESEARCH SUB-PROJECTS
 - CURRENT HARDWARE:
 - two Mark IV single cells
 - one Mark IV 12 cell stack
 - one Mark V 35 cell stack
 - one Mark V 5 cell high pressure stack

- TECHNICAL ACTIVITIES REFLECTED BY OUR PUBLIC DOMAIN PUBLICATIONS AND PRESENTATIONS

BALLARD HARDWARE USED AT THE ROYAL MILITARY COLLEGE OF CANADA

BALLARD STACK	# CELLS	SPECIAL FEATURES	ACTIVE SURFACE AREA (cm ² /cell)
Mark IV - 06	1	DuPont Nafion 117 membrane pure Pt catalyst	50.6
Mark IV - 07	1	DuPont Nafion 117 membrane pure Pt catalyst	50.6
Mark IV - 12	12	Dow Chemical membrane supplied catalyst	50.6
Mark V	35	5 kW system DuPont Nafion 117 membrane improved catalyst	230
Mark V D	5	High pressure system Ballard membrane improved catalyst	230

PEM Fuel Cell Activities

- Evaluation of Ballard Power Systems hardware delivered to the Department of National Defence (DND)
- Determination of performance limitations of Ballard hardware
- Assessment of contamination effects on Ballard hardware (eg. CO in the anode feed gas)
- Development of electrochemical performance models
- Development of steady-state and dynamic thermal models
- Systems integration and performance prediction of various applications

FUEL CELL PERFORMANCE MODEL

Output Voltage

$$V = E + \eta_{act} + \eta_{ir}$$

Thermodynamic potential

$$E = E_0^\circ - \frac{\Delta S^\circ}{nF}(T-298) + \frac{RT}{nF} \ln(p_{O_2}^* \cdot 2P_{H_2}^*)$$

Activation overvoltage

$$\eta_{act} = \xi_1 + \xi_2 T + \xi_3 T \ln(i) + \xi_4 T \ln(c_{O_2}^*)$$

Ohmic overvoltage

$$\eta_{ir} = -i (\xi_5 + \xi_6 T + \xi_7 i)$$

where the ξ 's represent parametric coefficients

GLOBE 90 FUEL CELL MODEL REVISIONS

$$\begin{aligned} V = & 1.229 - 0.85 * 10^{-3} (T - 298.15) \\ & + 4.3085 * 10^{-5} * T * [\ln(p_{H_2}^*) + \frac{1}{2} \ln(p_{O_2}^*)] \\ & + \xi_1 + \xi_2 T + \xi_3 T [\ln(c_{O_2}^*)] + \xi_4 T [\ln(i)] \\ & - i * [\xi_5 + \xi_6 T + \xi_7 i] \end{aligned}$$

for $138 \text{ ASF} \leq I \leq 388 \text{ ASF}$, $332 \text{ K} \leq T \leq 348 \text{ K}$, $0.15 \text{ atm} \leq p_{\text{air}} \leq 0.73 \text{ atm}$ (dry basis), and a constant p_{H_2} of 2.04 atm (dry basis)

Parametric Coefficients:

$$\xi_1 = -0.944$$

$$\xi_2 = 3.54 \times 10^{-3}$$

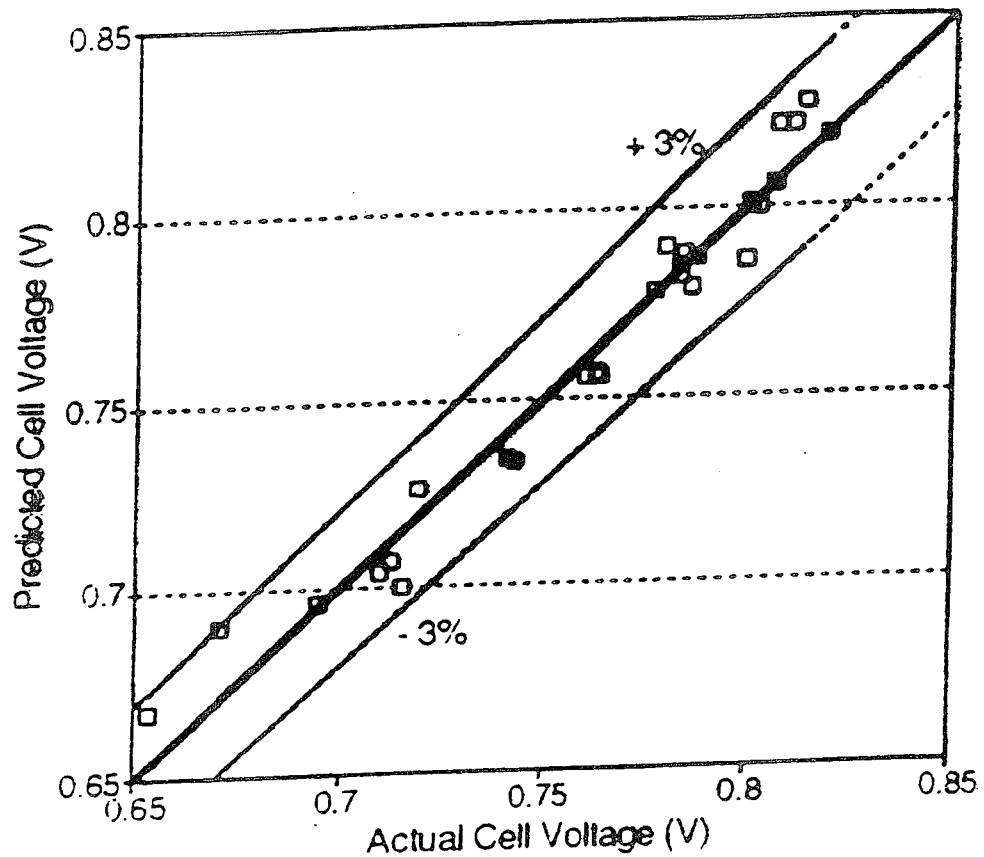
$$\xi_3 = 7.80 \times 10^{-5}$$

$$\xi_4 = -1.96 \times 10^{-4}$$

$$\xi_5 = 3.30 \times 10^{-3}$$

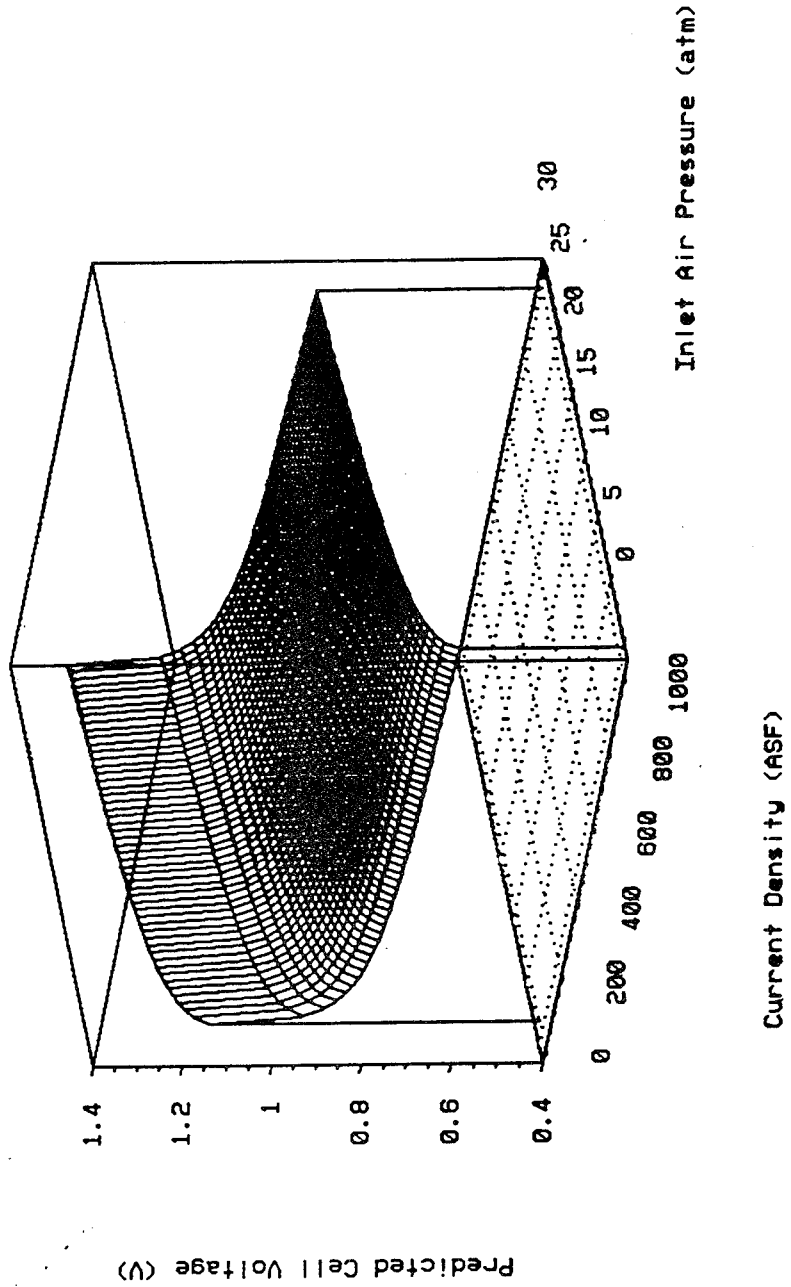
$$\xi_6 = -7.55 \times 10^{-6}$$

$$\xi_7 = 1.10 \times 10^{-6}$$



A parity plot of the experimental and predicted average cell voltage for the Globe 90

Predicted Voltage Surface Plot for the Ballard Globe 90 Stack at RMC



INCORPORATING THE EFFECTS OF CO INTO THE MODEL

Vogel (1975), Dhar (1987), and Wang (1992) proposed a relationship between the exchange current density, I° , and the fraction of surface coverage by CO at steady-state conditions, $\theta_{CO,s}$.

$$I_{CO}^{\circ} = I^{\circ}(1 - \theta_{CO,s})^2$$

The activation overvoltage, η_{act} , can be expressed in terms of exchange current density and the exchange current.

$$\eta_{act} = \frac{RT}{2F} \ln \left(\frac{I^{\circ}}{I} \right)$$

Combining the above equations yields

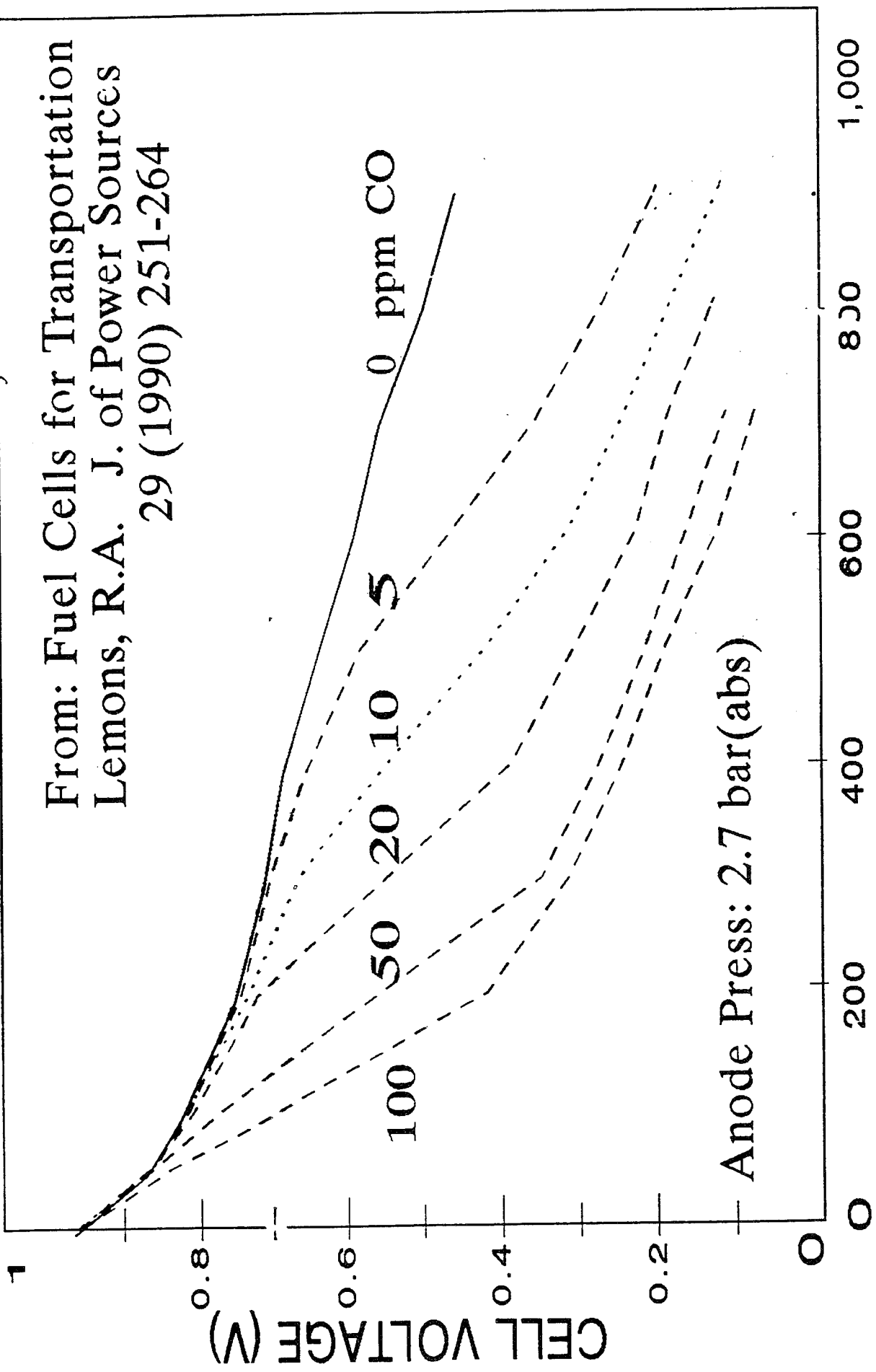
$$\eta_{act,CO} = \eta_{act} + \frac{RT}{F} \ln(1 - \theta_{CO,s})$$

Therefore, the output cell voltage contaminated with CO, V_{CO} , can be expressed as

$$V_{CO} = V + \frac{RT}{F} \ln(1 - \theta_{CO,s})$$

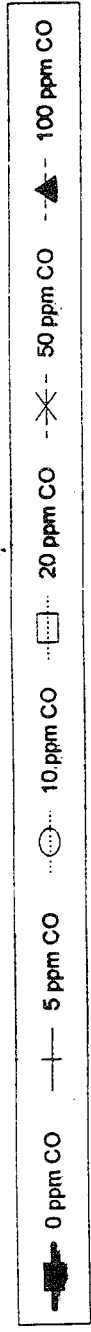
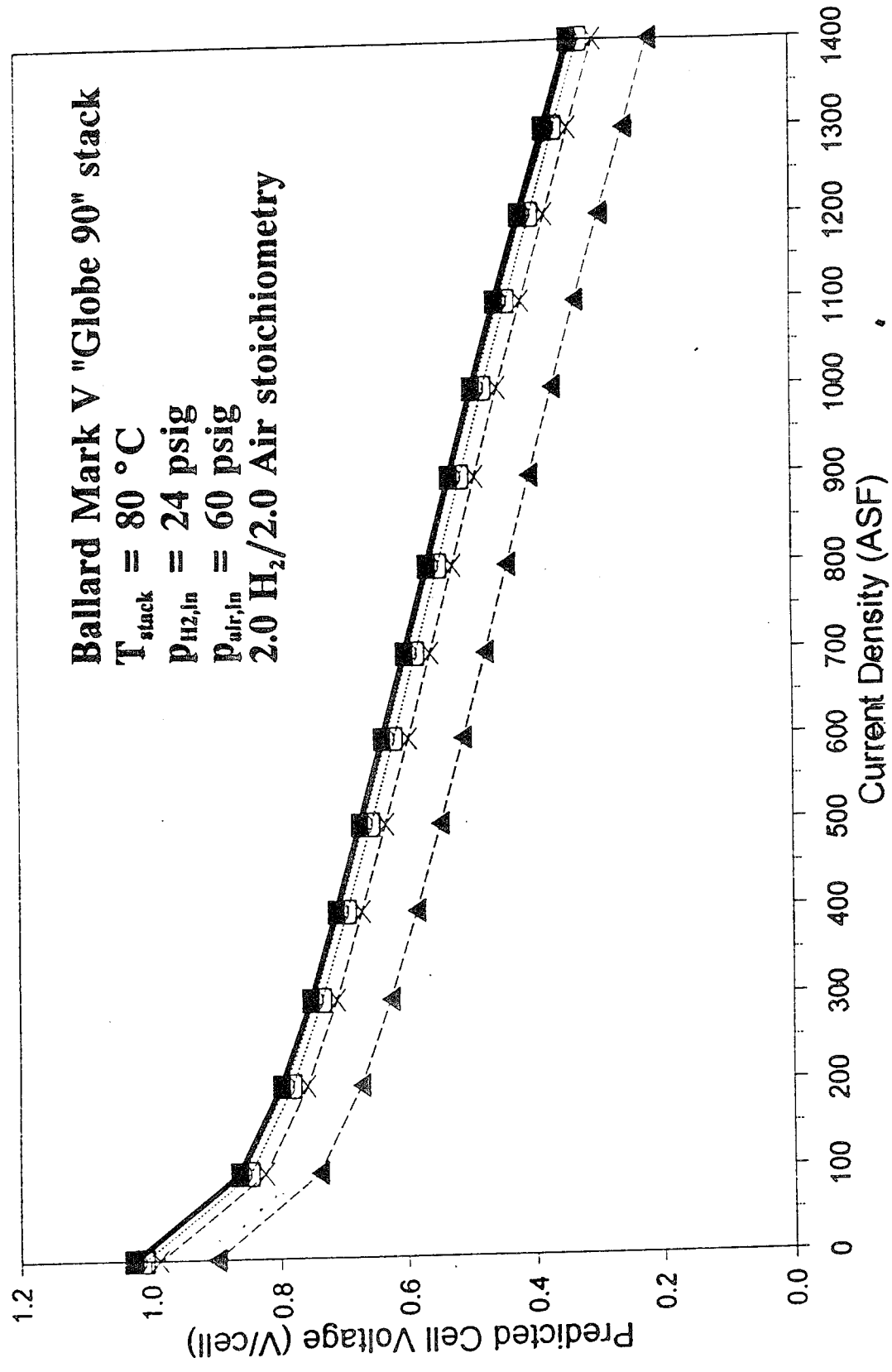
where V is the output cell voltage (without CO contamination) defined by our predicted model based on experimental data

From: Fuel Cells for Transportation
Lemons, R.A. J. of Power Sources
29 (1990) 251-264



CURRENT DENSITY (ASF)

Polarization Curves at Various CO Concentrations (θ_{CO} values based on Lemon's data)



RECENT DEVELOPMENTS

• PARAMETRIC PERFORMANCE MODEL DEVELOPED USING A COMBINATION OF MECHANISTIC AND EMPIRICAL MODELLING TECHNIQUES FOR THE BALLARD MARK IV SINGLE CELL

• MODEL APPLIED TO THE MARK V 35-CELL STACK WITH NEW PARAMETRIC COEFFICIENTS

• A PRELIMINARY PERFORMANCE MODEL INCORPORATING THE EFFECTS OF CARBON MONOXIDE CONTAMINATION ON PEM FUEL CELLS

FUTURE WORK

- **HIGH PRESSURE FUEL CELL OPERATION**

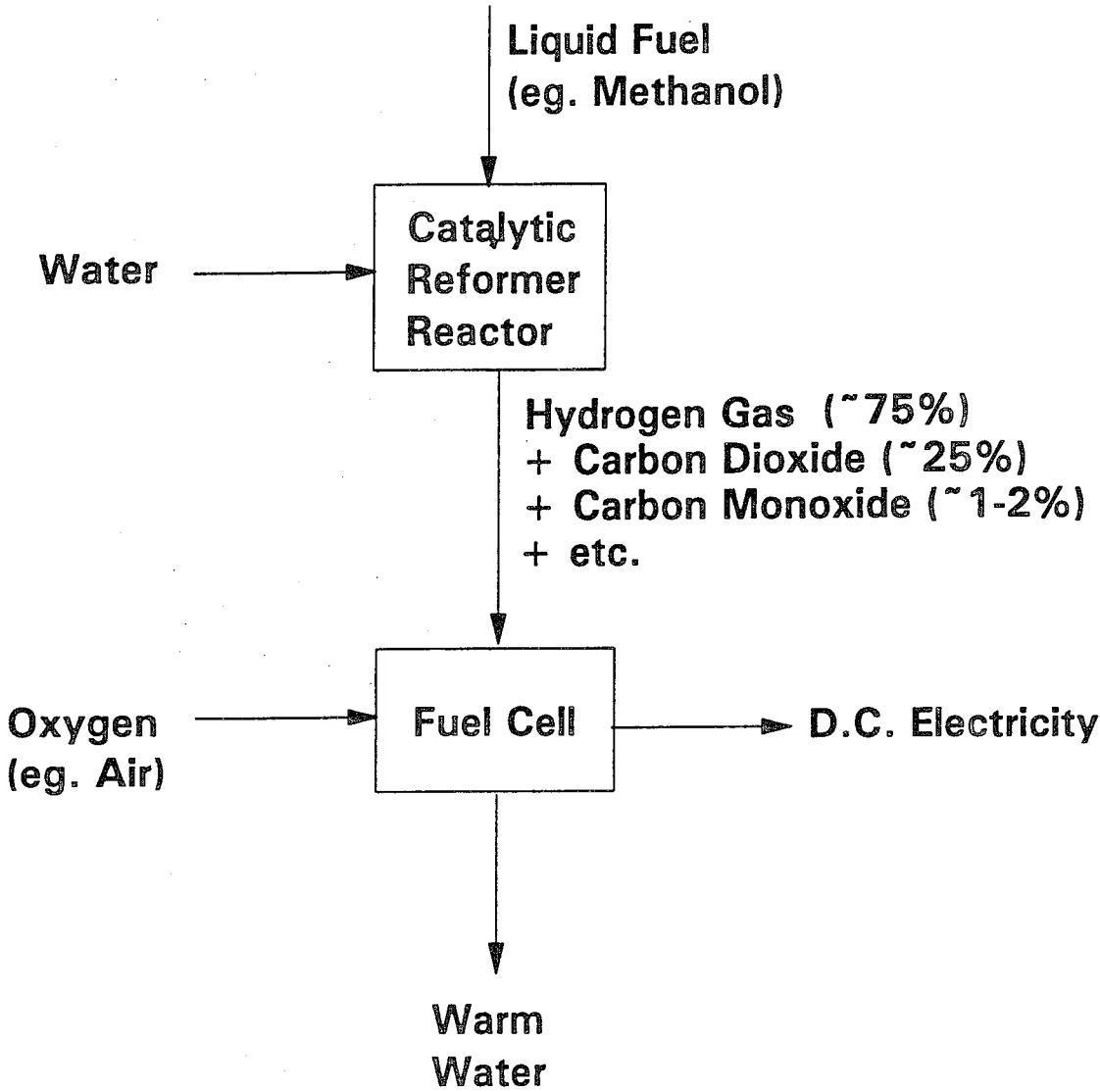
- **FUEL CELL OPERATION WITH CARBON MONOXIDE CONTAMINATION ON THE BALLARD MARK V D 5-CELL STACK**

- **FURTHER DEVELOPMENT ON THE PERFORMANCE MODEL TO INCORPORATE SURFACE CONDITIONS TO BETTER REPRESENT THE EFFECTS OF CONTAMINATION ON PEM FUEL CELLS**

Hydrogen Supply & Storage For Fuel Cells

- Supply of H₂ - rich gas from catalytic steam reforming of liquid fuels (primarily methanol)
- Purification of this "reformat gas" (eg.CO removal)
- Storage of 'pure' hydrogen
 - eg. • metal hydrides
 - chemical hydrides
 - microspheres
 - cryogenic adsorption
 - cryogenic liquid

FUEL CELL POWER GENERATING SYSTEM



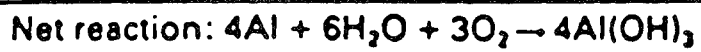
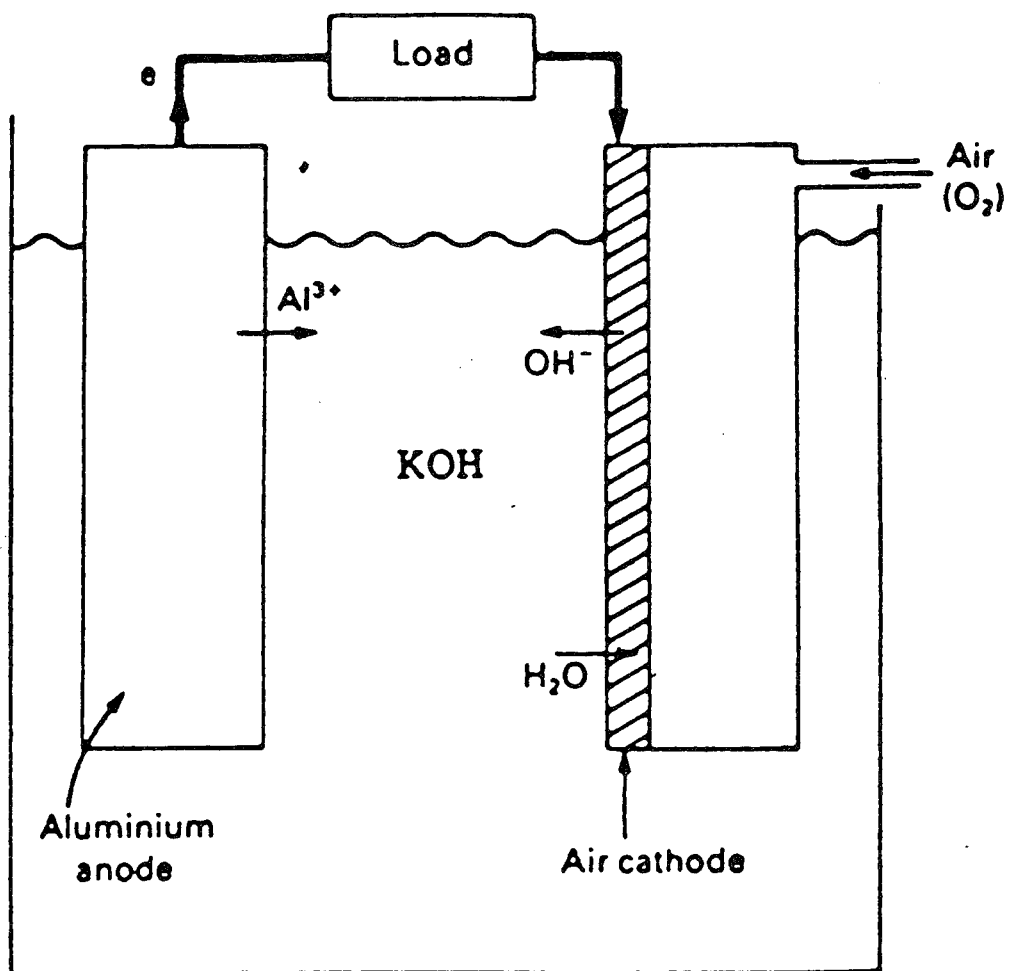
Oxygen Supply & Storage For Fuel Cells

- Liquid oxygen (LOX) systems
- Oxygen from catalytic decomposition of hydrogen peroxide
- Oxygen from air and 'oxygen enrichment'
- Oxygen from solid chemicals

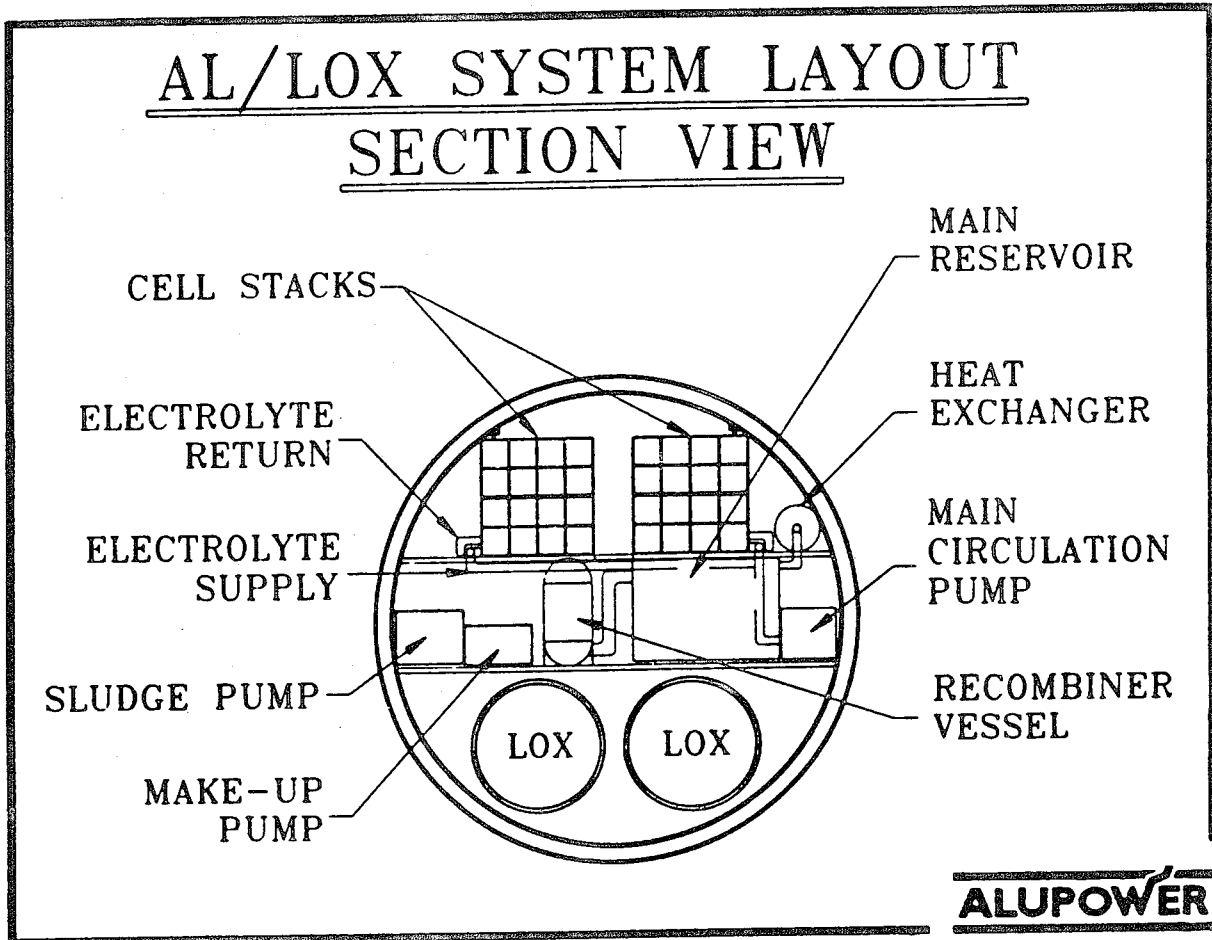
Non-PEM Fuel Cell Activities

- Evaluation of primary and secondary batteries
- Modelling of secondary battery charging & discharging
- Evaluation of aluminum / oxygen power cells (semi-fuel cells) (Alupower technology)
- Evaluation of small portable fuel cell power supplies (eg. LACEC Energy Systems alkaline cell / chemical hydride system)

SCHEMATIC OF ALUMINUM-AIR CELL



FACT SHEET ON AIR INDEPENDENT PROPULSION
FOR SUBMARINES



APPLICATION: Air-Independent Propulsion for Submarines

DESCRIPTION: Aluminum-Oxygen semi-fuel cell

BASELINE CAPACITY:

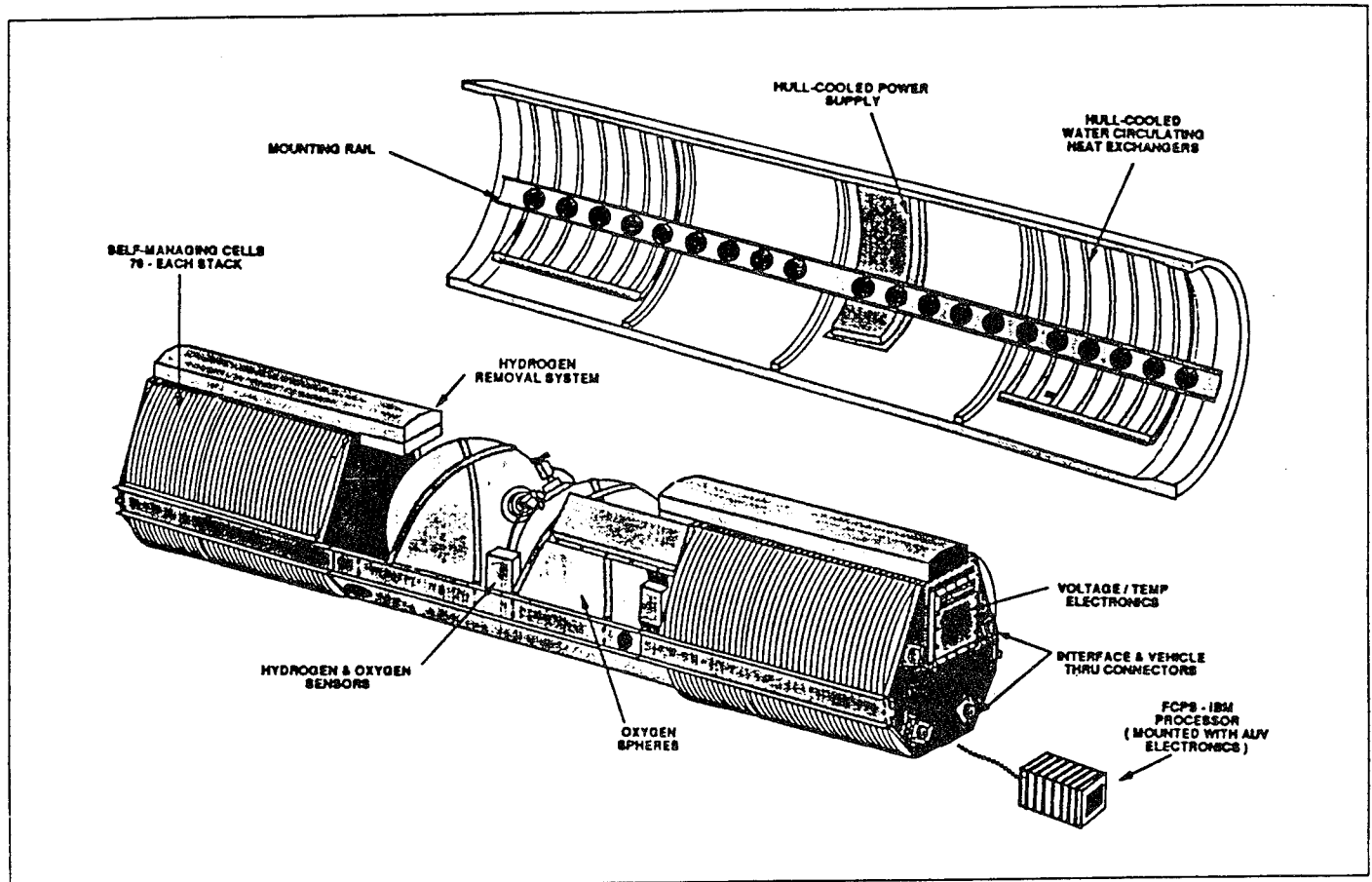
- 300 kW (Net) for 14 days submerged
- Nominal 440 V D.C.
- 390 kWh/m³ (including crew life support and all auxiliaries)
- Power Range: 50 to 800 kW

-- More --

NEWS FROM **ALUPOWER**

FACT SHEET ON ALUMINUM-OXYGEN FUEL CELL

POWER SYSTEM (FCPS) FOR UNDERWATER VEHICLES



XP-21 FUEL CELL POWER SYSTEM

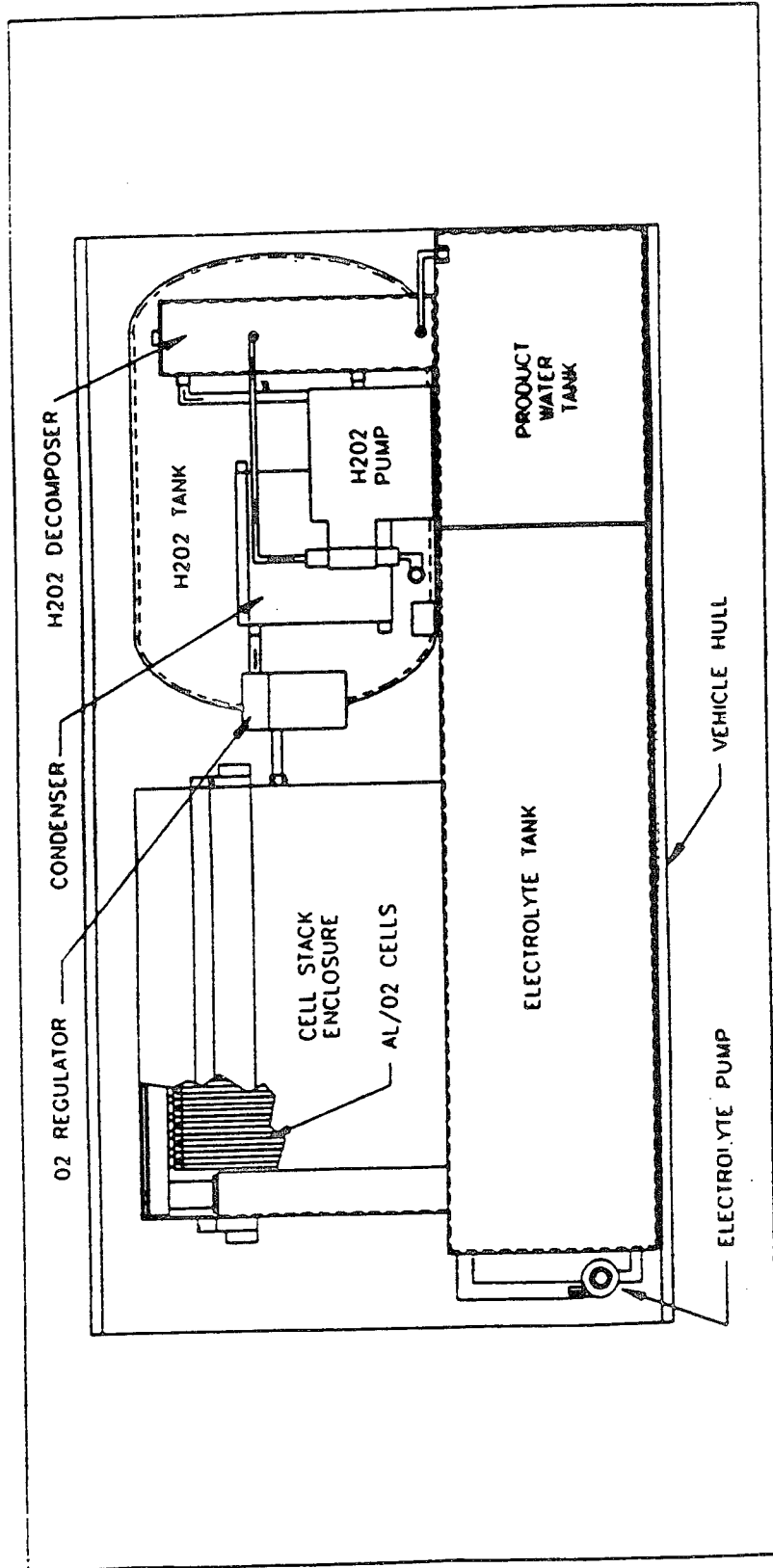
APPLICATION: Air-Independent Propulsion for Autonomous Underwater Vehicles

DESCRIPTION: Aluminum-Oxygen Semi-Fuel Cell

PERFORMANCE:

- Power - 2.5 kW (Net)
- Capacity - 100 kWh (Net)
- Voltage - 120 V Nominal
- Endurance - 40 Hours @ Full Power

-- More --





RMC

Royal Military College of Canada
Department of Chemistry & Chemical Engineering
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PUBLICATIONS AND PRESENTATIONS updated 15-08-94

(Fuel Cell and Hydrogen Supply Activities)

1994

"On Board Hydrogen Purification for Steam Reformer/PEM Fuel Cell Vehicle Power Plants", *Hydrogen Energy Progress X, Proceedings of the 10th World Hydrogen Energy Conference*, Cocoa Beach, Florida, USA, June 1994. J.C. Amphlett, R.F. Mann, and B.A. Peppley.

1993

"Temperature Programmed Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) of Methanol Decomposition on CuO/ZnO and CuO/Znd/Al₂O₃ Catalysts", 76th Canadian Society for Chemistry Conference, Sherbrooke, Que., June 1993. S.A. Colledan, J.C. Amphlett, K.A.M. Creber, R.F. Mann, B.A. Peppley, and D.M. Stokes.

"A Fuel Conditioning System for a Methanol-Fueled PEM Fuel Cell Power Generator", The First International Conference on New Energy Systems and Conversions, Yokohama, Japan, 613 - 618, June 1993. J.C. Amphlett, R.F. Mann, and B.A. Peppley.

"The Steam Reforming of Methanol: Mechanism and Kinetics Compared to the Methanol Synthesis Process", Third International Symposium on Natural Gas Conversion, Sydney, Australia, July 1993. J.C. Amphlett, R.F. Mann, and B.A. Peppley. (To be subsequently published by Elsevier in the Symposium Proceedings)

"Methanol, Diesel Fuel, and Ethanol as Liquid Sources of Hydrogen for P.E.M. Fuel Cells", *Proceedings of the 28th IECEC*, Atlanta, Georgia, Aug. 1993. J.C. Amphlett, R.D. Klassen, R.F. Mann, and B.A. Peppley.

"Performance Modelling of the Ballard Mark IV Solid Polymer Electrolyte Fuel Cell, Part I - Mechanistic Model Development", submitted to *J. of the Electrochem. Soc.*, June 1994. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, P.R. Roberge, and T.J. Harris.

"Performance Modelling of the Ballard Mark IV Solid Polymer Electrolyte Fuel Cell, Part II - Empirical Model Development", submitted to *J. of the Electrochem. Soc.*, June 1994. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, P.R. Roberge, and T.J. Harris.

"Development and Application of a P.E.M. Fuel Cell Performance Model", *Proceedings of the 28th IECEC*, Atlanta, Georgia, Aug. 1993. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"The Effect of Carbon Monoxide Contamination on Anode Efficiency in PEM Fuel Cells", *Prep. pap. 206th American Chemical Society National Meeting* 38, 4 (1993) 1477-1482. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, P.R. Roberge, and A. Rodrigues.

"Parametric Modelling of the Performance of a 5 kW Proton Exchange Membrane Fuel Cell Stack", *J. Power Sources* 49 (1994) 349-356. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, P.R. Roberge, and A. Rodrigues.

"Recycle Reactor Model for Highly Dispersed Flow in Catalytic Fixed Bed Reactors", 43rd Canadian Chemical Engineering Conference, Ottawa, Ontario, Oct. 1993. B.A. Peppley, J.C. Amphlett, and R.F. Mann.

1992

"An Experimental Design for Determining the Optimum Method of Catalyst Preparation for Low Temperature Methanol Steam Reforming", *Proceedings of 12th Canadian Symposium on Catalysis*, Banff, Alberta, May 1992 (*Progress in Catalysis*, 73, 343-349, Elsevier Science Publishers, Amsterdam, 1992). J.C. Amphlett, K.A.M. Creber, J.M. Davis, R.F. Mann, B.A. Peppley, and D.M. Stokes.

"Hydrogen Production by Steam Reforming Methanol for Polymer Electrolyte Fuel Cells", *Proceedings of the 9th World Hydrogen Energy Conference*, Paris, June 1992, pp. 1541-1550. J.C. Amphlett, J.M. Davis, R.F. Mann, B.A. Peppley, and D.M. Stokes. (subsequently refereed and published in the *International Journal of Hydrogen Energy*).

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"Methanol as a Source of Hydrogen - Rich Gas for a Solid Polymer Fuel Cell Power System", 75th Canadian Chemical Conference, Edmonton, Alberta, June 1992. J.C. Amphlett, K.A.M. Creber, J.M. Davis, R.F. Mann, B.A. Peppley, and D.M. Stokes.

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fo
"Optimization of a Fuel Cell/Battery Hybrid Underwater Power Source", 182nd Meeting. The Electrochemical Society, Toronto, October 1992. J.C. Amphlett, R.M. Baumert, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"Toward an Optimized Hydrogen Supply System for a Methanol Fueled PEM Fuel Cell Power Generator", 182nd Meeting. The Electrochemical Society, Toronto, October 1992. J.C. Amphlett, J.M. Davis, R.F. Mann, and B.A. Peppley.

"Catalytic Steam Reforming of Methanol for a PEM Fuel-Cell Power Plant", 42nd Canadian Chemical Engineering Conference, Toronto, October 1992. J.C. Amphlett, J.M. Davis, R.F. Mann, B.A. Peppley, and D.M. Stokes.

"Methanol Fuels and PEM Fuel Cells for Vehicle Transportation", 42nd Canadian Chemical Engineering Conference, Toronto, October 1992. J.C. Amphlett, R.M. Baumert, M. Farahani, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"Reconciliation of the Kinetics of Methanol Decomposition on Girdler G66B Catalyst as Obtained by Amphlett et al. *Can. J. Chem. Eng.* 63, 605-611 (1985)". Note, *Can. J. Chem. Eng.* 70, 207-208 (1992). J.C. Amphlett and R.F. Mann.

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"The Operation of a Solid Polymer Fuel Cell: A Parametric Model", *Proceedings of the 26th Intersociety Energy Conversion Engineering Conference*, (Paper 910055), Boston, Mass., Aug. 1991. J.C. Amphlett, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"Some Design Considerations for a Catalytic Methanol Steam Reformer for a PEM Fuel Cell Power Generating System", *Proceedings of the 26th Intersociety Energy Conversion Engineering Conference*, (Paper 910056), Boston, Mass., Aug. 1991. J.C. Amphlett, R.F. Mann, B.A. Peppley, and D.M. Stokes.

"Performance Analysis of the Ballard Solid Polymer Electrolyte Fuel Cell", 42nd Meeting of the International Society of Electrochemistry, Montreux, Switzerland, Aug. 25- 30, 1991. J.C. Amphlett, A.V. Anantaraman, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"Operating Characteristics of a Solid Polymer Fuel Cell", *Proceedings of Project Hydrogen '91*, Independence Missouri, Sept. 1991. J.C. Amphlett, M. Farahani, R.F. Mann, B.A. Peppley, and P.R. Roberge.

"Computer Simulation of a Catalytic (Methanol Reforming) Hydrogen Generator for a Portable Fuel-Cell Power Plant", 41st Canadian Chemical Engineering Conference, Vancouver, B.C., Oct. 6-9, 1991. J.C. Amphlett, T.J. Harris, R.F. Mann, B.A. Peppley, and P.R. Roberge.

1990

"Preparation and Characterization of Three Methanol Steam Reforming Catalysts Based on Copper-Magnesia with Additions of Manganese, Chromium and Vanadium Oxides", 11th Canadian Symposium on Catalysis, Halifax, July 1990. J.C. Amphlett, R.F. Mann, B.A. Peppley, and D.M. Stokes.

1989

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"Solid Polymer Electrolyte Fuel Cells: Independent Test Evaluation of Ballard Technologies Corporation Fuel Cells", 72nd Canadian Chemical Conference, Victoria, B.C., June 1989. J.C. Amphlett, A.V. Anantaraman, R.F. Mann, B.A. Peppley, and P.R. Roberge.

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1987

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"Hydrogen Production by the Catalytic Steam Reforming of Methanol - Part 2: Kinetics of Methanol Decomposition using Girdler G66B Catalyst", *Can. J. Chem. Eng.* **63**, 605-611 (1985). J.C. Amphlett, M.J. Evans, R.F. Mann, and R.D. Weir.

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"The Steam Reforming of Methanol on CuO/MgO Catalysts, *Proceedings of 2nd International Symposium on Hydrogen Produced from Renewable Energy*, 91-100, Cocoa Beach, Florida, October 1985. J.C. Amphlett and R.F. Mann.

1981

"Hydrogen Production by the Catalytic Steam Reforming of Methanol - Part 1: Thermodynamics, *Can. J. Chem. Eng.* **59**, 720-727 (1981). J.C. Amphlett, M.J. Evans, R.A. Jones, R.F. Mann, and R.D. Weir.

ELECTRODEPOSITION OF NAFION ONTO ELECTRODE STRUCTURES

Bruce J. Tatarchuk
Chemical Engineering Department
&
Space Power Institute
Auburn University, AL 36849
November 1, 1994

The author, in collaboration with others shown, is conducting research on the electrodeposition of Nafion onto electrode structures as part of a larger piece of sponsored research. The author gratefully acknowledges the support for this work under Army Contract #DAAL0392G0205-2.

Technical Approach. The technical approach is to extend the electrode reaction zone away from the PEM and into three dimensions, thereby gaining improved performance of the electrode device in the particular application under study. Important attributes of the "3-D Reaction Zone" involve accessibility and/or transport issues which are driven by the contacting of the "extended electrode-PEM microstructure" to one or more of the following: the catalyst, the reactants and its products, the electrons, and the interface between the PEM/SPE and the electrode. There are some issues which are not addressed in this topic as presently described -- the technical work does not address proton conductivities, membrane permeation, and catalyst deactivation, among others. The design approach is described by expanding from a 2-D model (shortest distance between catalyst/electrode and PEM is straight line) to a 3-D design in which electrodes are fabricated from fibers, not particulates.

Several recent publications cover the subjects of design, electrochemical properties, and characterization of composite fuel cell electrodes, in more detail, i. e., scientific results of the authors' studies of the electrochemical reaction of oxygen at the electrocoated Nafion-modified metal carbon composite and platinum electrodes, as well as the use of electrocatalytic metal-carbon composite electrodes for SPE fuel cells are described.

Experimental Procedure. The experimental procedure was described as the preparation of like electrode samples for further processing using coating, dipping, electrocoating, and optimizing electrocoating procedures. The proprietary process used to fabricate the electrode samples ensures that they are of suitable (similar if not almost identical) thickness, physical composition, uniformity of metal/carbon fibers and void volume, and possess similar electrical and chemical characteristics prior to coating, dipping, etc. Results of the Nafion loading with respect to time were reported. Potential versus current curves were compared to show the effects of electrocoating versus dipping on the electrical characteristics. Nafion packing and cost of materials were also studied/reported.

Results. The results indicated that electrocoating does work and can be controlled (and repeated with reliable accuracy). The research team is using preferential placement of Nafion as a technique to maximize performance versus cost, follow design contours, maximize the benefit/volume ratio, attain more uniform and self-correcting deposition procedures, to take advantage of the fact that differential rates allow placement and high rates form external films, to use in tandem with hot-pressing to reduce SPE membrane thickness and resistance, to improve mechanical toughness, and to decrease electrode/SPE interfacial resistance.

November 1, 1994

Prospector VII: Small Fuel Cells for Portable Power

Electrodeposition of Nafion onto Electrode Structures

B. Tatarchuk
G. Swain
H. Xu
J. Wang
R. Killough

Department of Chemical Engineering, and
the Space Power Institute
Auburn University, Alabama

Acknowledgments:

Army - DAAL0392G0205 - 2

Approach is to Extend Electrode Reaction Zone away from the PEM and into Three Dimensions for Improved Performance

Important Attributes of a 3-D Reaction Zone Involve Accessibility/Transport Issues Driven by Contacting of an Extended Electrode-PEM Microstructure to the:

- a. Catalyst**
 - Effectiveness Factor
 - Dispersion (Both Forms)

- b. Reactants/Products**
 - Heat transfer
 - Mass Transfer
 - Water Balance (pore size, hydrophobicity, control)

- c. Electrons**
 - Intra-Electrode Conductivity/Thickness
 - Contact to the Collector

- d. Interface between PEM/SPE and Electrode**

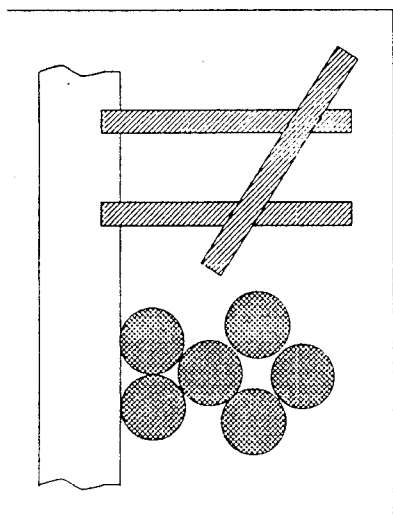
Have Not Included

- Proton Conductivities
- Membrane Permeation
- Catalyst Deactivation
- Etc.

Design Approach

2-D Model: Shortest Distance between Two Points, e.g., Catalyst/Electrode and PEM, is a Straight Line

3-D Implications: Fabricate Electrodes from Fibers Not Particulates



Fabricate Added Intra-Electrode PEM Bridges along Straight Paths, Don't Use Much

Eliminate Binders by Combining Binder and Collector Functions

Use Electrocoating Methods

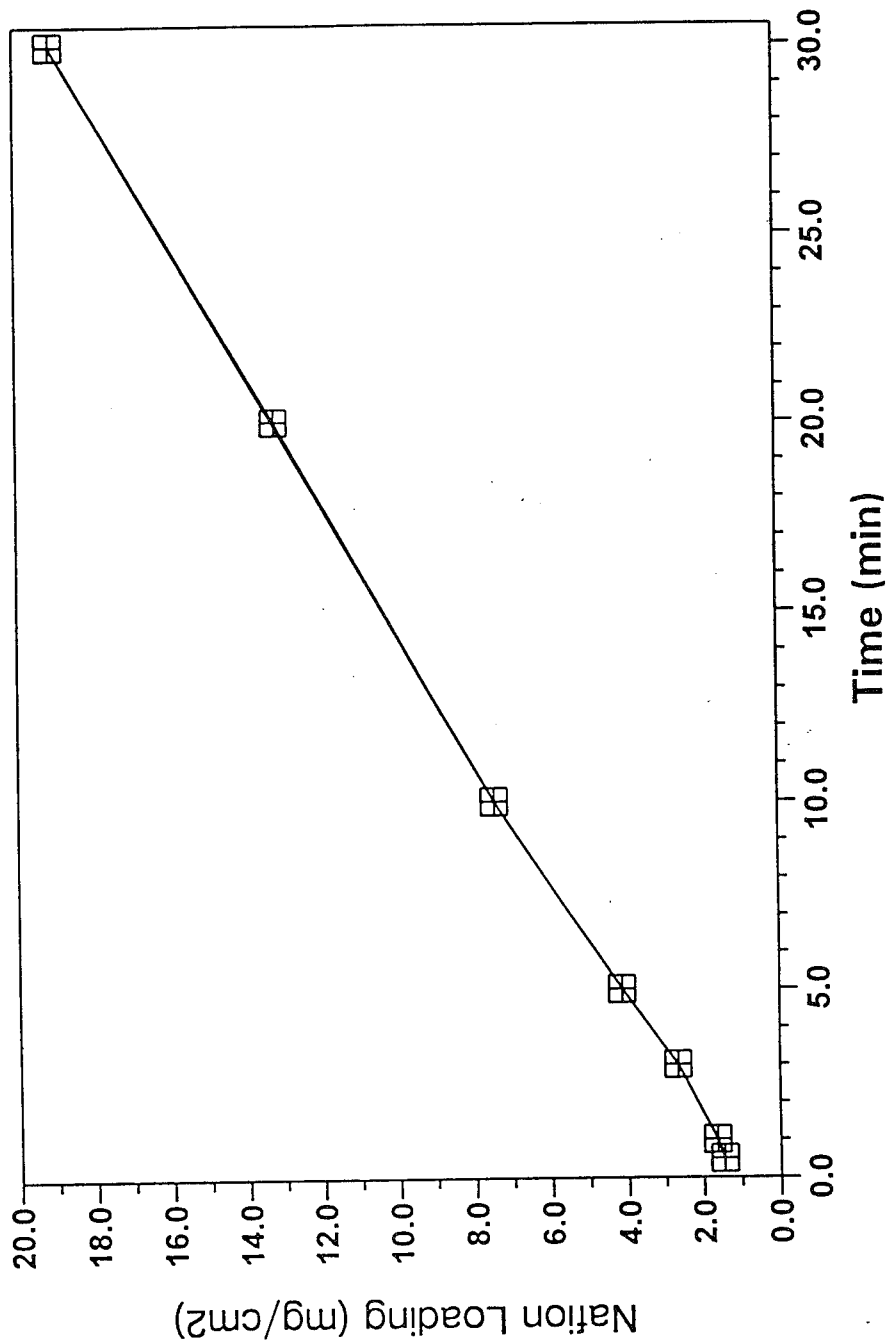
e.g., "Electrochemical Reaction of Oxygen at Electrocoated Nafion-Modified Metal Carbon Composite and Platinum Electrodes," G.M. Swain and B. J. Tatarchuk, *J. Electrochemical Soc.*, 140 (4), 1026-1033 (1993)

"Electrocatalytic Metal-Carbon Composite Electrodes for SPE Fuel Cells," H. Xu, R. Killough, C. Marrion, S. Ahn, and B. J. Tatarchuk, 1994 Annual Meeting of the AIChE, San Francisco, CA, November 1994.

Experimental Procedure

- a. Prepare series of identical metal-carbon composite electrodes with: ~ 1 g of 2 μm Ni fibers, ~ 1 g of bundled 2 μm activated carbon fibers, and ~ 0.5 g of cellulose fibers; per 200 cm^2 of preform.
- b. After sintering average wt. per 1.6 cm^2 electrode (@ 140 μm thickness) is 11.86 mg \pm 1.5%
- c. Check various coating procedures, dip versus electrocoating, optimize electrocoating.

Nafion loading vs. electrocoating time. Samples were electrocoated at a potential of 2.0 V in 5 wt. % Nafion solution of lower aliphatic alcohols and water.



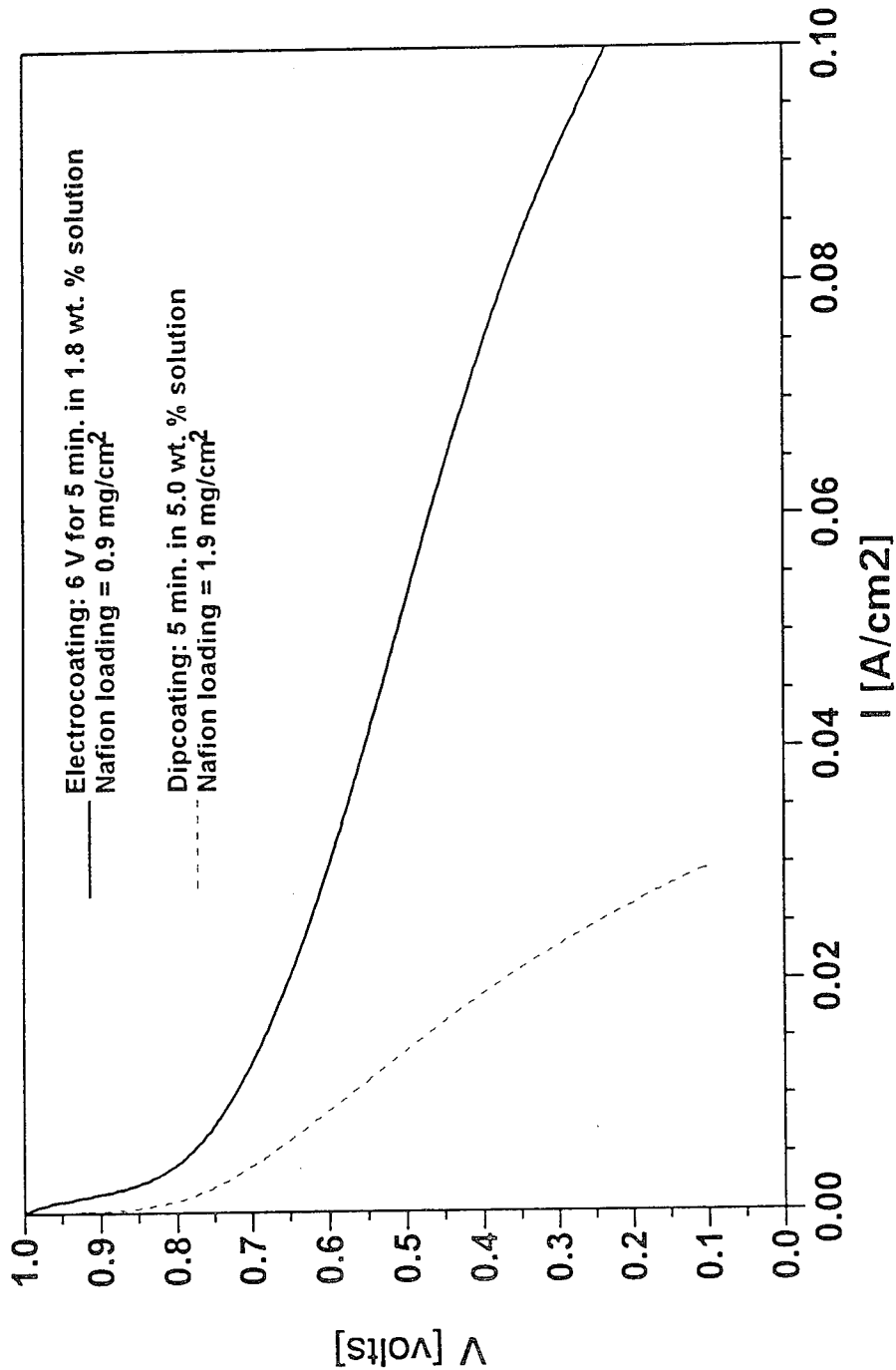
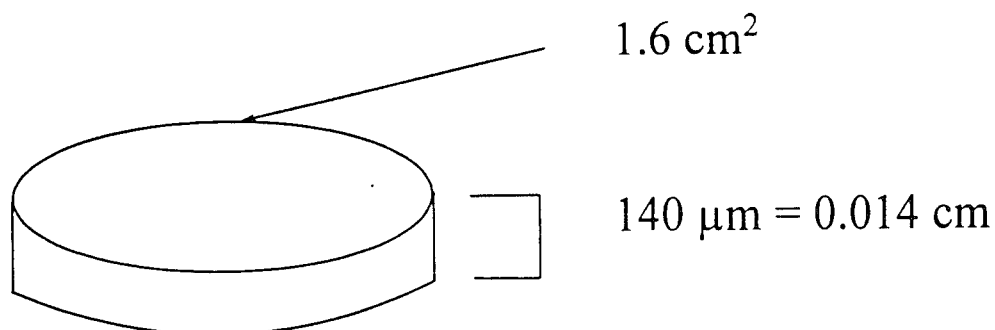


Figure 4. Potential vs. current curves of H₂/O₂ SPE fuel cell using composite electrodes with different Nafion impregnation methods at room temperature and atmospheric pressure. A Nafion 117 membrane is used as polymer electrolyte. The composition of the electrode is 1.0 / 1.0 / 0.5 (Ni / C / Cellulose) in the preform.

Calculation of Nafion Packing



$$\text{Total Electrode Volume} = 0.0224 \text{ cm}^3$$

$$\text{Electrode Weight} = 11.86 \text{ mg or}$$

$$5.93 \text{ mg Ni @ } 8 \text{ g/cm}^3 = 7.4 \times 10^{-4} \text{ cm}^3$$

$$+ 5.93 \text{ mg C @ } 2 \text{ g/cm}^3 = 2.97 \times 10^{-3} \text{ cm}^3$$

$$\text{Total Volume of Solids} \quad \underline{\hspace{1.5cm}} \quad 3.71 \times 10^{-3} \text{ cm}^3$$

$$\text{Electrode Void Volume} = 0.0224 - 3.71 \times 10^{-3} = 0.01869 \text{ cm}^3$$

$$\text{Electrode Void Volume} = \frac{0.0224 - 3.71 \times 10^{-3}}{0.0224} \times 100\%$$

$$= 83.4\%$$

$$\text{Nafion Filling} = 0.01869 \text{ cm}^3/\text{electrode} \div 1.6 \text{ cm}^2/\text{electrode}$$

$$\times \rho_{\text{Nafion EC}} @ 0.7 \text{ g/cm}^3$$

$$\cong 8.2 \text{ mg/cm}^2 \text{ of electrode}$$

Cost of Materials for Electrode Fabrication

$$\frac{\$600}{18 \text{ L}} \mid \frac{1 \text{ L}}{1000 \text{ ml}} \mid \frac{1 \text{ ml}}{0.8923 \text{ g}} \mid \frac{1 \text{ g soln}}{0.05 \text{ g Nafion}} = \frac{\$7.47}{\text{g Nafion}}$$

$$\frac{\$7.47}{\text{g Nafion}} \mid \frac{0.0082 \text{ g Nafion}}{\text{cm}^2} = \frac{\$0.06 \text{ of Nafion}}{\text{cm}^2} \quad (\text{complete filling})$$

$$\frac{\$450}{\text{Troy Oz Pt}} \mid \frac{1 \text{ Tr. Oz.}}{31.1 \text{ g}} = \frac{\$14.47}{\text{g Pt}}$$

$$\frac{11.86 \text{ mg electrode}}{1.6 \text{ cm}^2} \mid \frac{1 \text{ g}}{1000 \text{ mg}} \mid \frac{0.01 \text{ g Pt}}{\text{g electrode}} \mid \frac{\$14.47}{\text{g Pt}} = \frac{\$0.001 \text{ of Pt}}{\text{cm}^2}$$

$$\frac{11.86 \text{ mg electrode}}{1.6 \text{ cm}^2} \mid \frac{1 \text{ g}}{1000 \text{ mg}} \mid \frac{0.1 \text{ g Pt}}{\text{g electrode}} \mid \frac{\$14.47}{\text{g Pt}} = \frac{\$0.01 \text{ of Pt}}{\text{cm}^2}$$

$$\frac{\$20}{\text{lb Ni}} \mid \frac{1 \text{ lb}}{454 \text{ g}} \mid \frac{11.86 \text{ mg electrode}}{1.6 \text{ cm}^2} \mid \frac{1 \text{ g}}{1000 \text{ mg}} \mid \frac{\$14.47}{\text{g Pt}} = \frac{\$1.64 \times 10^{-4} \text{ of Ni}}{\text{cm}^2}$$

Carbon ~ same as Ni

Man - Tech and PEM Film Not Included.

Summary and Implications

Electrocoating works and can be controlled

Preferential Placement of Nafion to:

- a. Maximize performance versus cost**
- b. Follow design contours**
- c. Maximize benefit/volume ratio**
- d. Uniform and self-correcting deposition process**
- e. Differential rates allow placement, high rates form external films**
- f. Use in tandem with hot pressing to reduce SPE membrane thickness and resistance, improve mechanical toughness**
- g. Decrease electrode/SPE interfacial resistance**

Prospector VII: Small, Portable Fuel Cells

October 31 - November 3, 1994
Durham, North Carolina

ZERO CROSSOVER DIRECT METHANOL FUEL CELLS

Chemistry Department
Illinois Institute of Technology

Eugene Smotkin

Wayne Huang

Cong Pu

Funding for this work is provided by the
Army Research Office

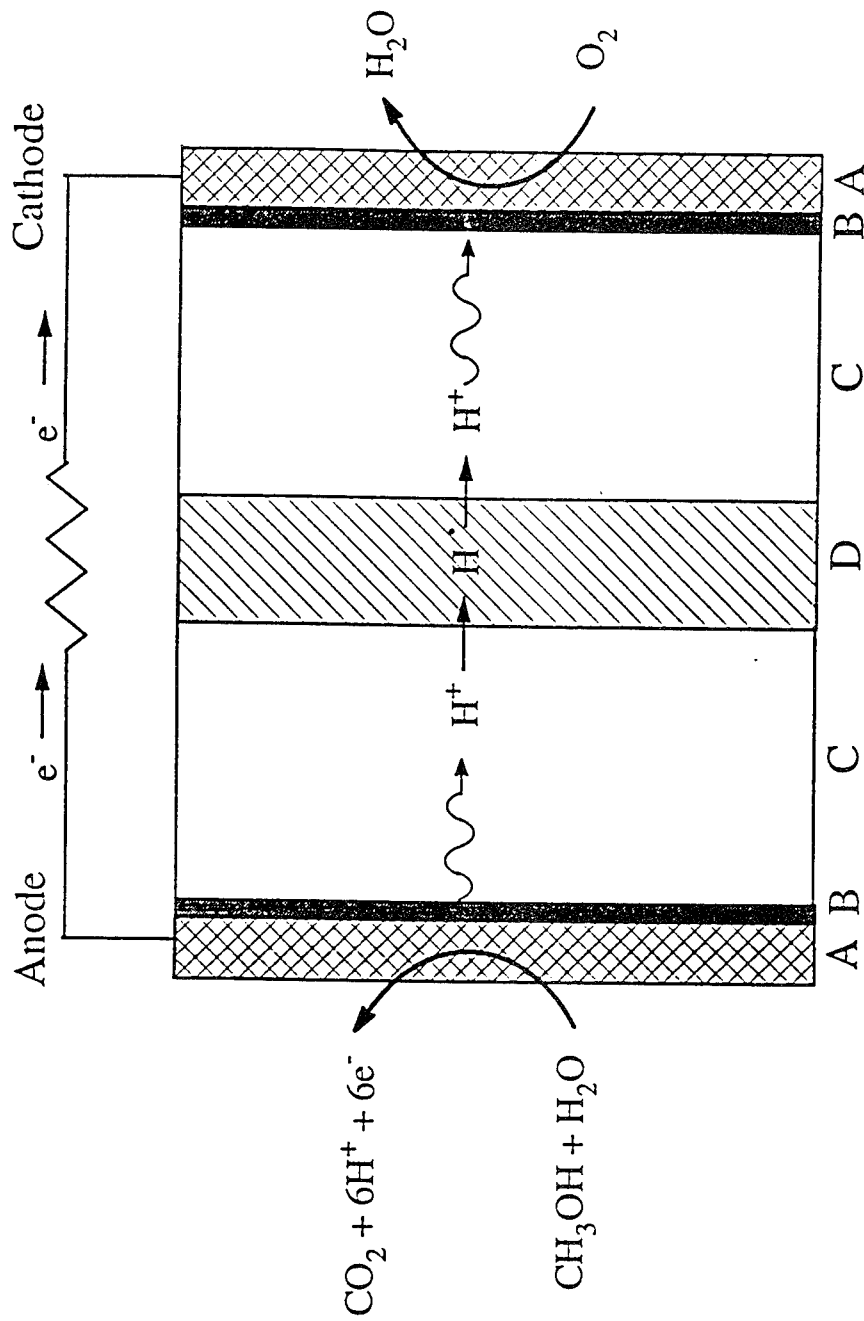
Figure Captions

Figure 1. Schematic diagram of the new composite electrolyte system containing methanol impermeable protonic conductor (MIPC). The porous gas diffusion electrodes are prepared from carbon cloth, and are coated on one side with a layer of electrocatalyst comprising 20% Pt on Vulcan XC-72 (E-Tek, Inc., Natick, MA). The data in this report were obtained with Nafion 115 serving as the PPEI, and a 25 μm thick palladium film as the MIPC.

Figure 2. I-V curves of a conventional fuel cell with two Nafion TM 115 membranes as the electrolyte. Curve A was obtained with H_2 as the fuel. Curves B and C were obtained 1 and 7 hr., respectively, after adding 12% methanol to the H_2 humidifier.

Figure 3. I-V curves of differently configured fuel cells containing the palladium MIPC sandwiched between two Nafion 115 membranes, which serve as the PPEIs. Curve A was obtained with a ca. 30 \AA Pt-modified palladium foil (25 μm) sandwiched between two Nafion 115 membranes, and H_2 as the fuel. Curve B was obtained 20 hr. after adding 10% methanol to the H_2 humidifier, using the same electrolyte system as in Curve A. Curve C was obtained using unmodified palladium foil sandwiched between two Nafion 115 membranes and H_2 as the fuel.

Figure 4. Effect of electrodeposition of Pt on Pd films. I-V curve is superimposed over Figure 2 H_2/O_2 performance curves.



- A Porous Gas Diffusion Electrode
- B Electrocatalyst Layer
- C Proton Permeable Electronic Insulator
- D Methanol Impermeable Palladium Membrane

Fig. 1

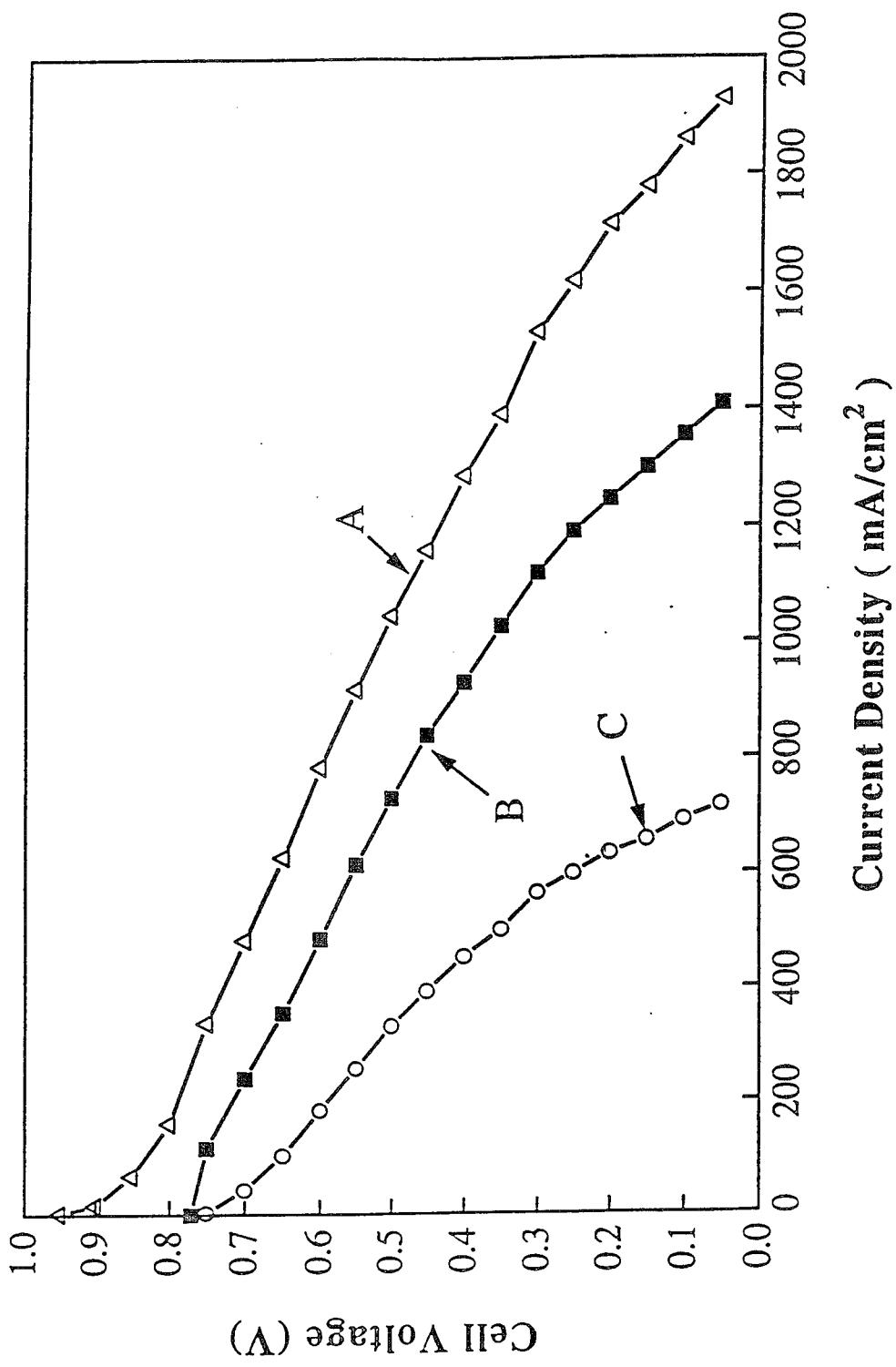


Fig. 2

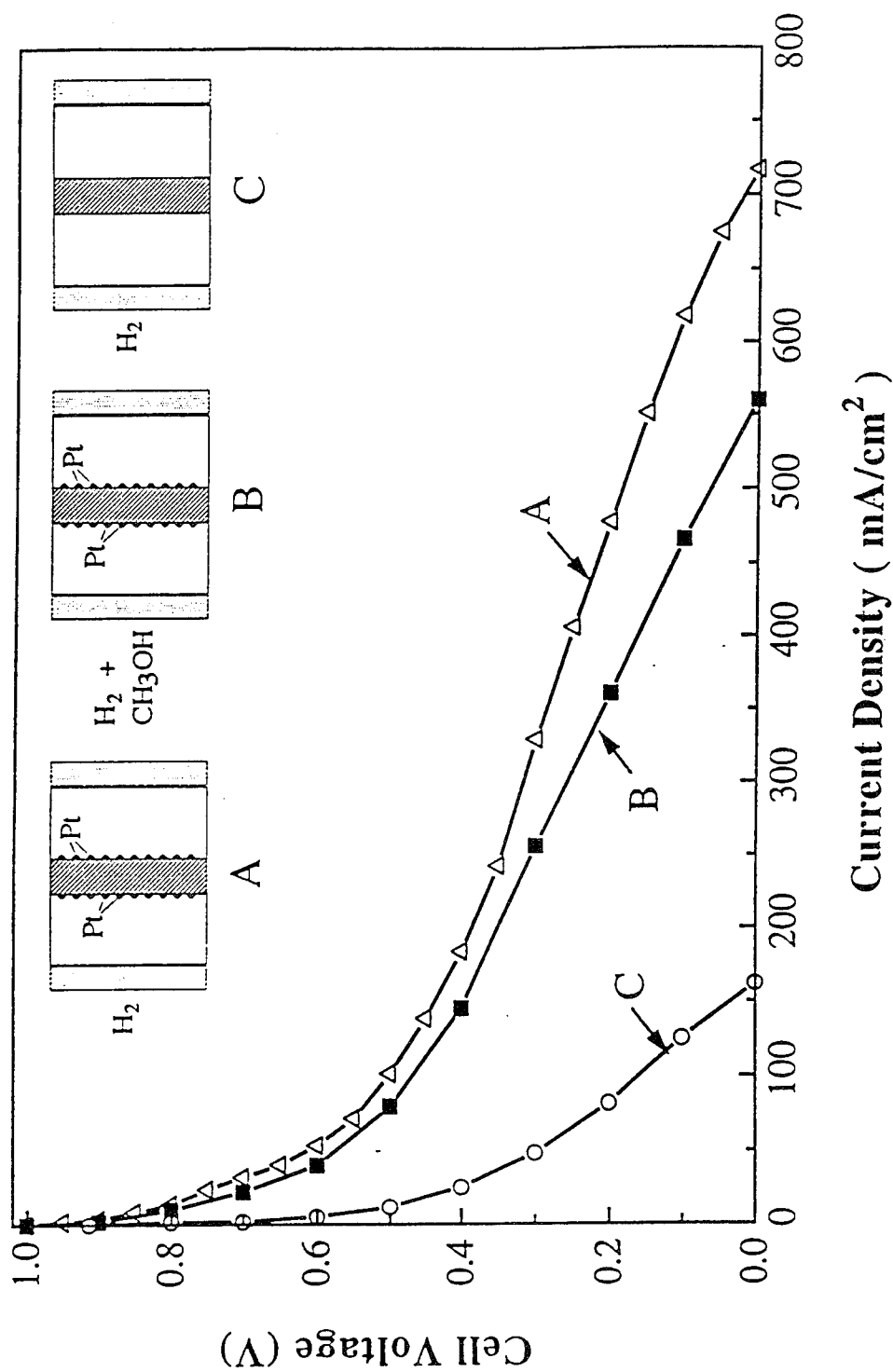
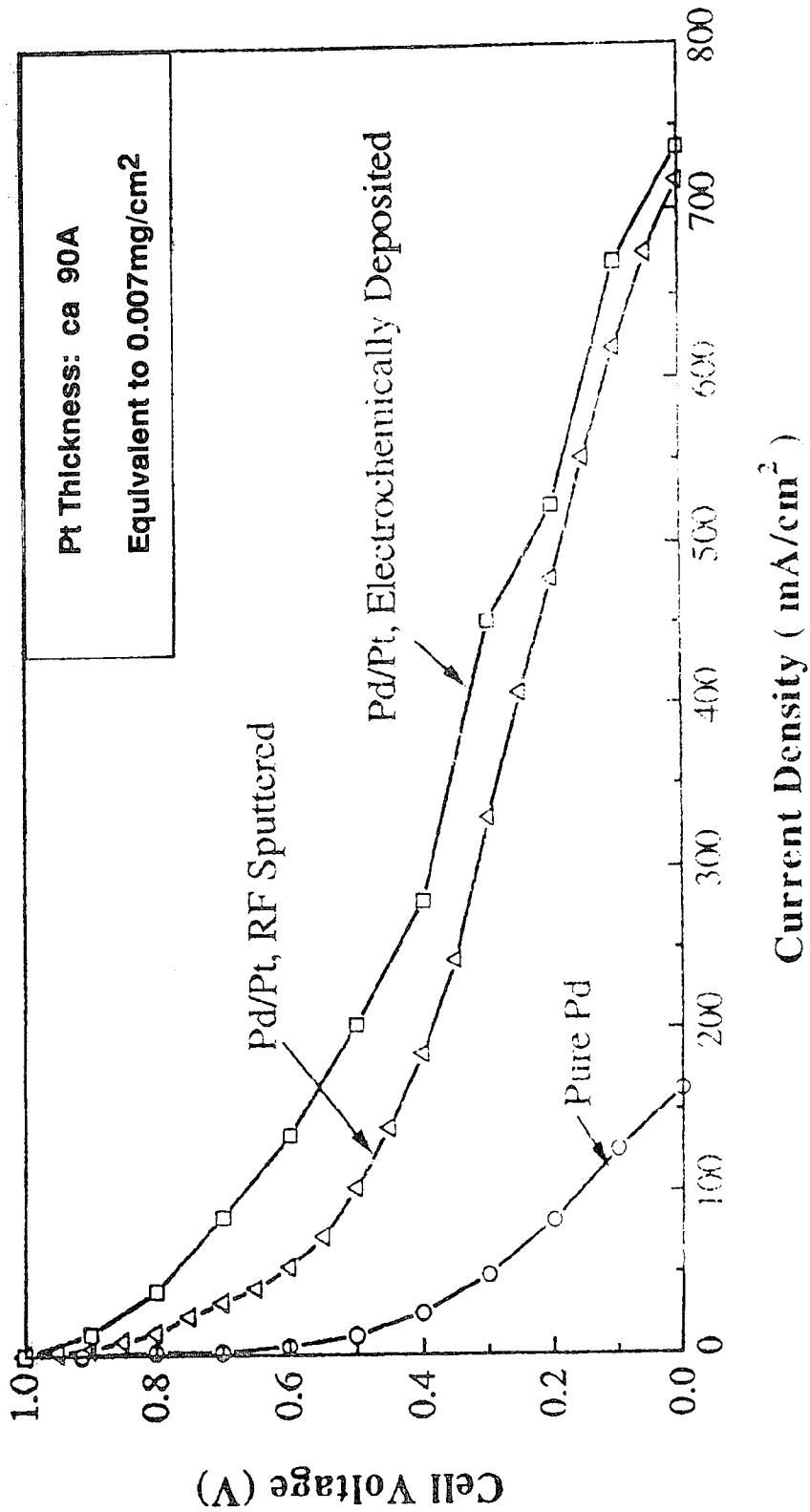


Fig. 3



Temperature: Cell 100 C, Sparger - 105 C, Both
Back Pressure: 30 psig Both
Gas Flow Rate: H₂ = 500 SCCM; O₂ = 1000 SCCM

Fig. 4

TECHNOLOGY UPDATE III

TECHNOLOGY UPDATE SESSION III

Chairman: Dr. Bob Hamlen, ARL

The session began with introductory remarks by the session chairman. He commented on the need to reflect upon such technical issues as low temperature options, heat transfer and cooling challenges, methods of producing and storing hydrogen as a fuel, and development of a hybrid power source design where portions of the Soldier System power would be provided by non-fuel cell means.

- "Low Temp Solid Oxide Fuel Cells at Argonne," Dr. Mike Krumpelt, Argonne Laboratory
- "Heat Transfer and Cooling Challenges in Fuel Cell Development," Mr. Dave Bloomfield, Analytic Power, Inc
- "Field Production of H₂: Wet or Dry Manufacture from Military Transport Fuels," Dr. Bill Peters, MIT
- "Update on Battery Workshop (Prospector III)," Dr. Bob Guenther, ARO

The remainder of this section is a compilation of the technical material as submitted to the Workshop organizers. At the conclusion of this session, Dr. Dick Paur, charged the members of the Workshop on behalf of the Board of Directors. In his charge, he made the following points:

Charge to Participants. The individual Working Groups are charged with finding the best development paths and technology solutions for small, portable FC systems. "Small," for the purpose of this Workshop discussion, is defined as "less than 1 kW at the upper end and as small as practical at the lower end." *We want to emphasize that the complete power system has to be carried, so the energy density of complete systems is what really counts.* Additionally, Group I (Applications) should assess the SOA, assess whether it was feasible to go to smaller units to attempt to replace batteries, assess commercial markets and issues, and address system issues of stack integration, attitude sensitivity, shelf life, start-up time, disposal, and safety. Group II (Alternatives) should identify alternatives to FC in small size ranges, assess alternative FC technologies for systems under 1 kW, summarize advantages and disadvantages of alternate technologies when compared to fuel cells, and identify self-contained options and size range of application (assess the marketability of fuel cells) in military and commercial markets. Group III (Research Issues) is charged with developing a consensus of the leading technical "show-stoppers" limiting the rapid development of small FC units, prioritizing these leading technologies, identifying SOA performance and goals for future work, summarizing the potential strategies for near-term improvements and "breakthroughs," and identifying the basic scientific support needs for FC improvements.

With this set of questions and challenges, Working Group members were to seek out new ideas, identify the technological impediments, and secure comparisons of traditional and non-traditional technologies. The group was divided into three main areas as follows:

- Civilian and Military Fuel Cell **Applications** for Commercial Products and Specific Missions
- Alternative Fuel Cell **Technologies**
- **Key Research Issues** Pacing Development - Definitions & Priorities

The summaries of those discussions, and the respective Group briefings to the entire body of participants, are included in these Proceedings.

WORKSHOP OBJECTIVES

The **Workshop objectives**, as determined by the Board of Directors, were to:

- Assess the state-of-the-art of fuel cells and fuel cell systems by characterizing innovative fuel cell conversion techniques and determining their applicability to DOD and civil power systems;
- Identify the key research issues pacing the development of (or limiting full development of) efficient, high power fuel cells/systems with acceptable lifetimes;
- Identify the major limiting factors which must be addressed as part of overall fuel cell/system design for man-portable applications;
- Prioritize and clearly define research issues, indicating the impact if successful;
- Provide research team milestones to assure significant improvements in fuel cell technology over near-term and long-term development programs;
- Identify operational/environmental constraints such as materials, signatures, mass, and pollution which influence the applications or improvements envisioned; and
- Establish scaling laws, and wherever possible, use those laws to compare results with other methods of powering DOD and civil systems.

To accomplish these goals, a group of distinguished scientists and engineers, active in the field, were invited to present, in several overview talks, current perceptions of the state-of-the-art in fuel cell power systems. Each speaker was to address key issues, operational and environmental constraints, and potential military and commercial applications for small, portable fuel cell systems from their own particular vantage points in DOD agencies, academic institutions, and research laboratories.

Summary of Presentation

"Direct Methanol Solid Oxide Fuel Cells"

by

Rajiv Doshi and Michael Krumpelt
Argonne National Laboratory

A new type of solid oxide fuel cell is being developed at Argonne National Laboratory (ANL) for operation on air and methanol. This Direct Methanol Solid Oxide Fuel Cell (DMSOFC) is designed to operate in the temperature range 450-500°C without external reforming. The DMSOFC is made of all ceramic materials alternating with a bipolar plate which can be metal. There are no liquids in any of the components.

There are several inherent design advantages to the DMSOFC that are outlined here:

- 1) CO tolerant anodes (no noble metals)
- 2) Ambient pressure operation
- 3) No methanol cross-over
- 4) Water generated on anode side (no water management required)
- 5) Potentially compatible with other reformed fuels without CO removal
- 6) High chemical and thermal stability of components

This technology is being pursued at Imperial College, U.K., and at Argonne National Laboratory in the U.S. Imperial College has exhibited a performance of 150 mA/cm² at 0.7 V. Argonne has obtained an open circuit voltage of 0.97 V at 450°C.

The electrolyte is a doped ceria oxygen ion conductor. Cerium oxide was considered not stable enough in a high temperature solid oxide fuel cell (SOFC) operating above 800°C due to reduction and subsequent onset of electronic conductivity in reducing atmospheres. We have shown that at the reduced temperatures of this new fuel cell operating on methanol, cerium oxide reduction does not occur and, the material remains an ionic conductor in both air and methanol. The advantage of using doped ceria is the significantly higher oxygen ion conductivity compared to stabilized zirconia which is currently used in high temperature SOFCs. In fact the conductivity of doped ceria is almost an order of magnitude higher than stabilized zirconia at 450°C.

Anodes and cathodes for this new type of SOFC still need to be improved to obtain good performance. Both electrode materials need to be good ionic and electronic conductors, and need to be formulated to match the thermal expansion coefficient of the electrolyte.

To facilitate rapid start-up, the materials need to be reinforced for thermal stress tolerance. When fully developed, a SOFC operating at 450°C and tolerant to carbon monoxide may be applicable for power systems running on logistics fuel.

Krum

Direct Methanol Solid Oxide Fuel Cells

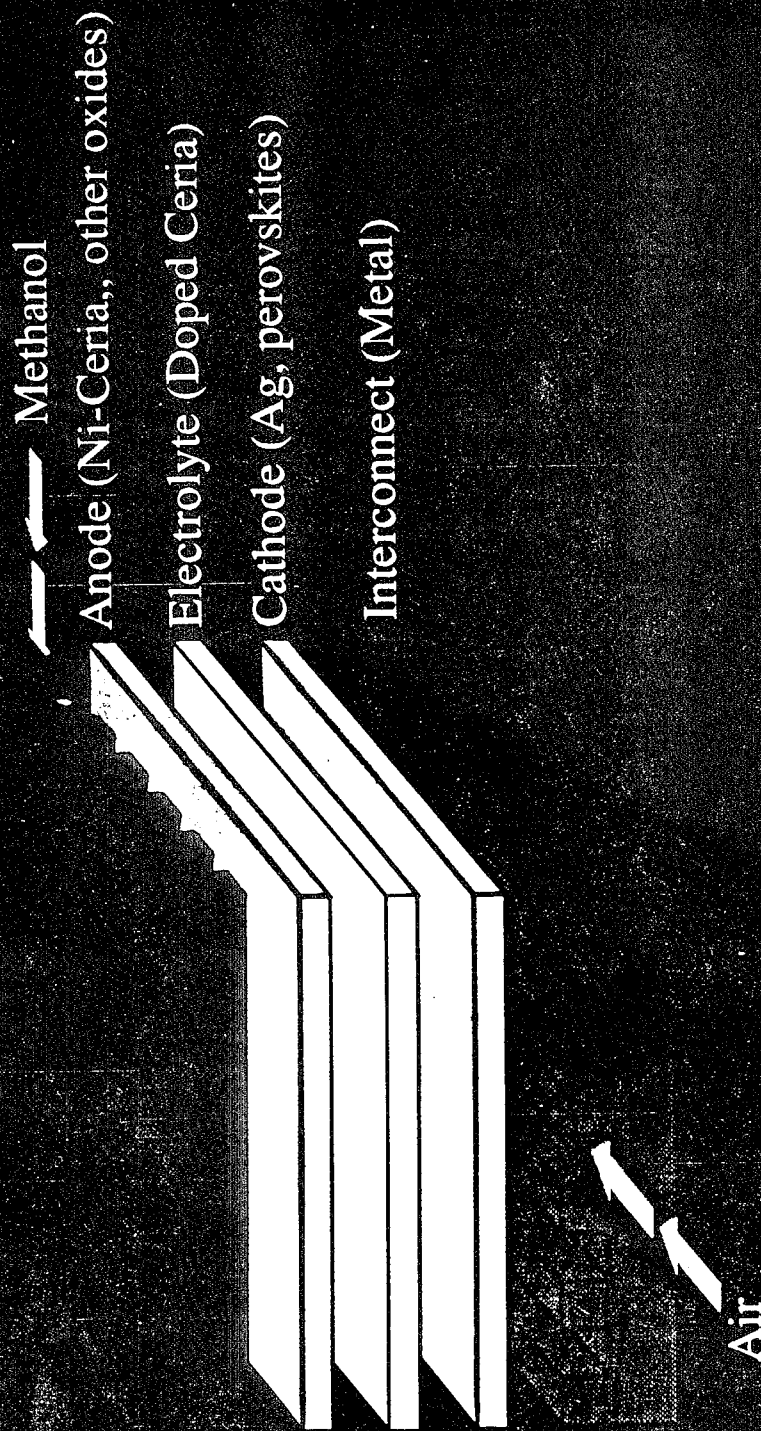
R. Doshi

M. Krumpelt

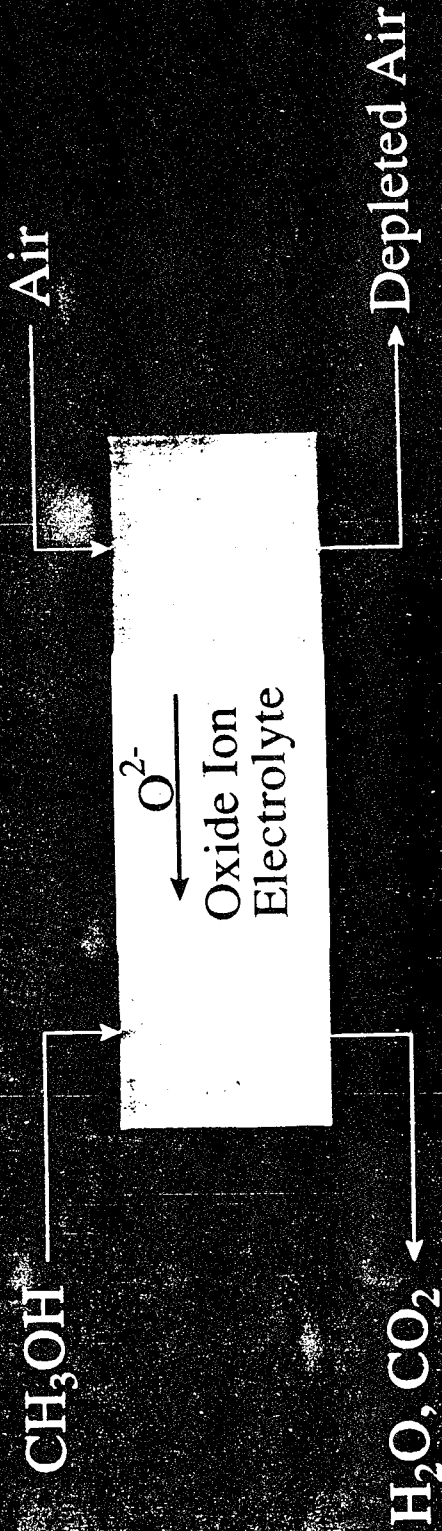


November 1, 1994

Planar DMISOFC



Reactions in a DMSOFC



anode:

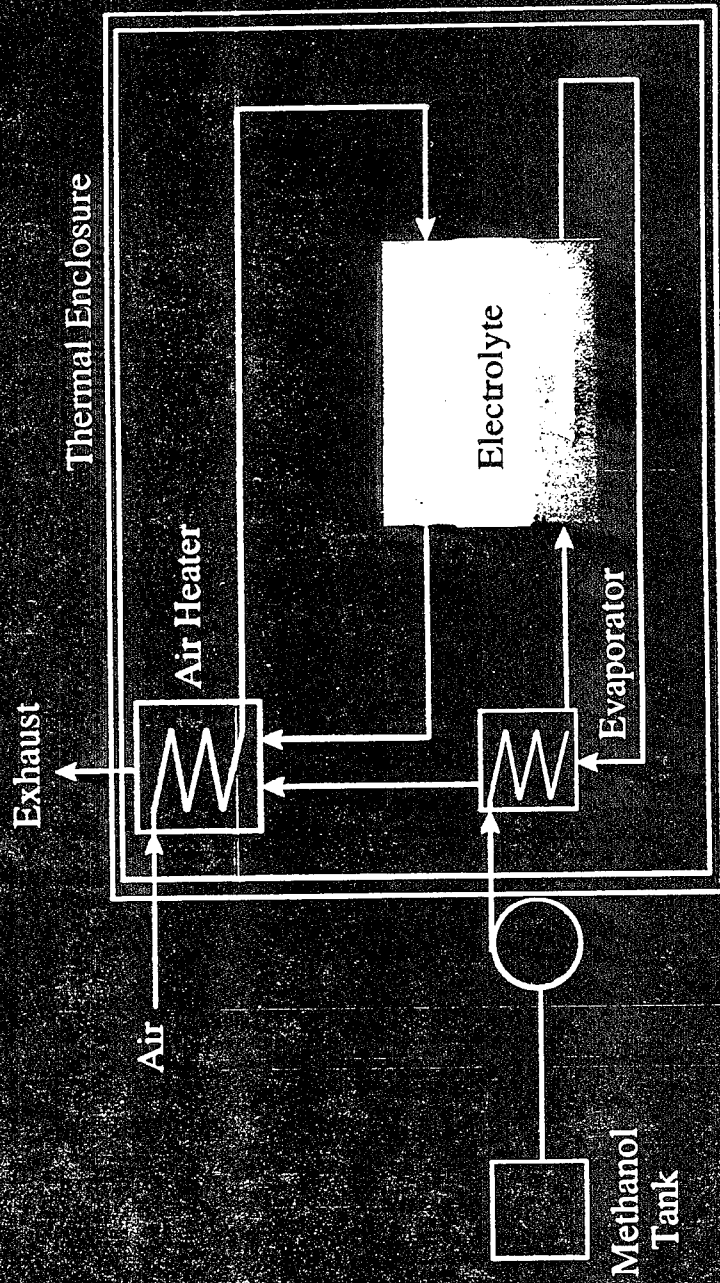


cathode:

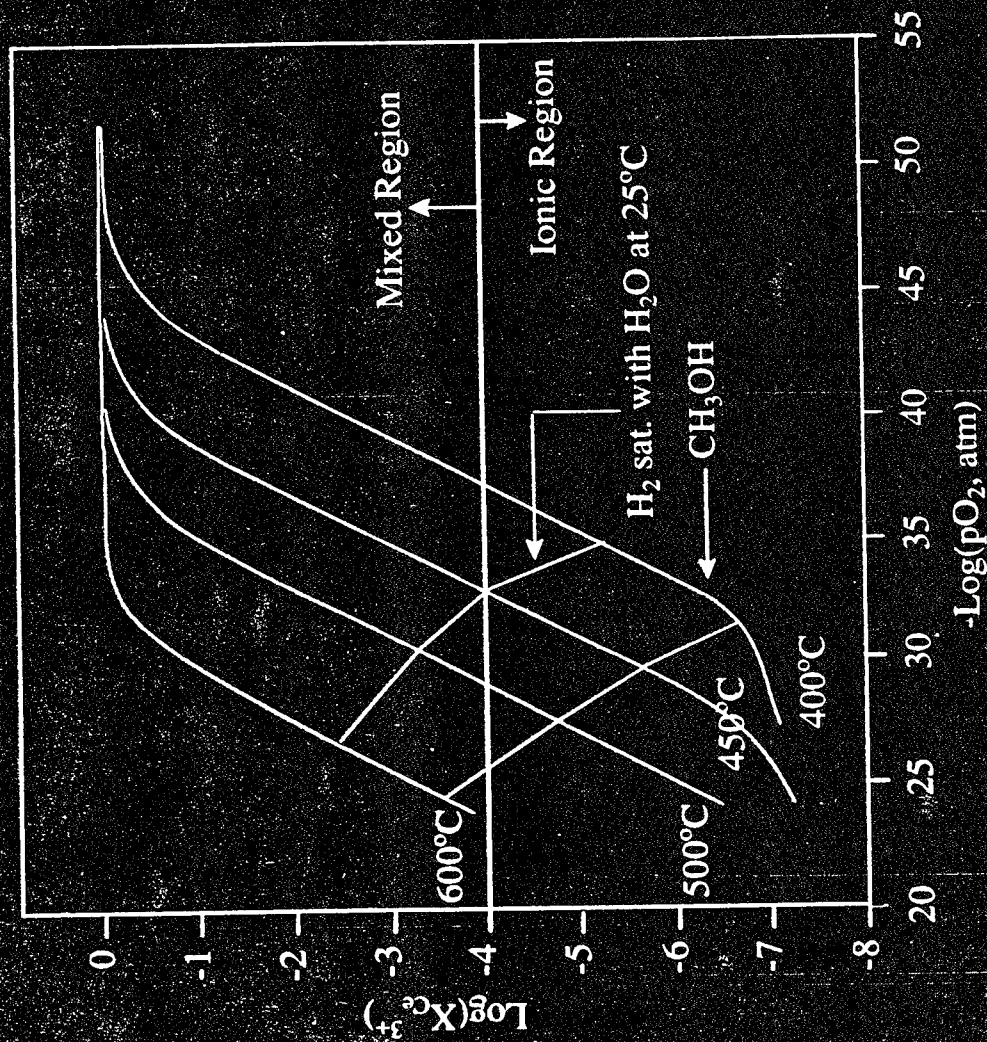


DMSOFC system schematic

A DMSOFC system with three major components

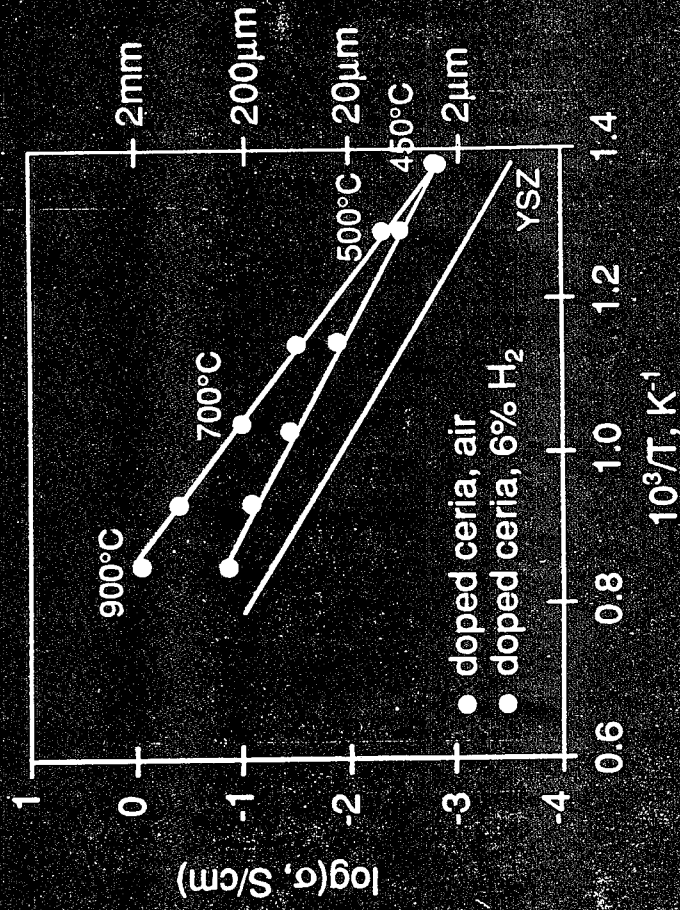


Electrolyte stability in methanol

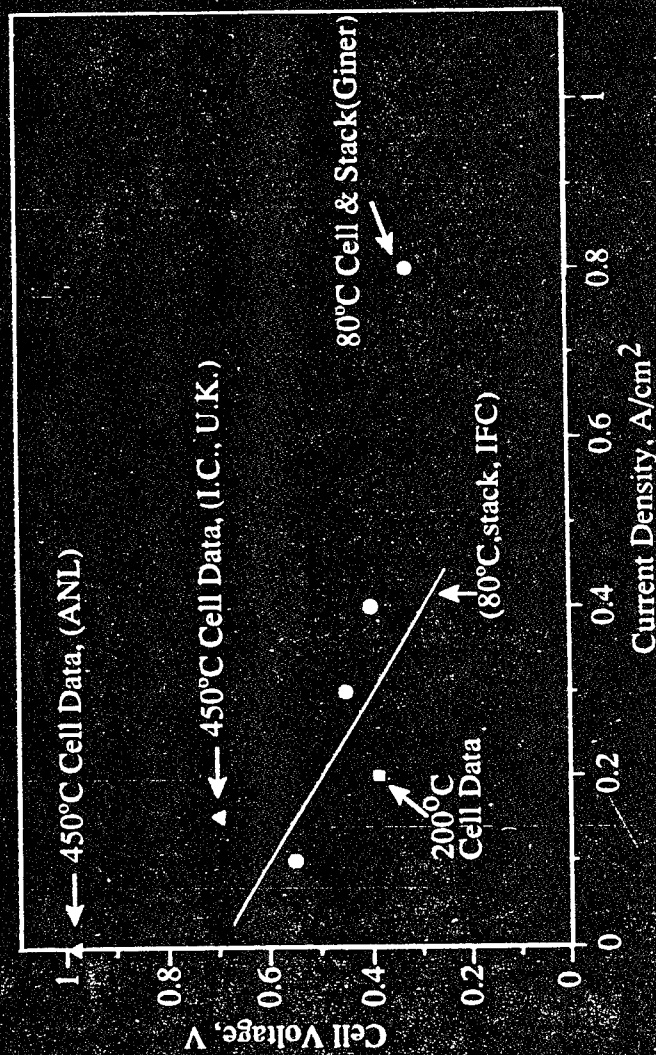


Conductivity of Electrolyte

Doped ceria is a much better ionic conductor than YSZ



Methanol Fuel Cells - Polarization Data



The DM SOFC still needs some work

- Imperial College has shown a performance of 150 mA/cm^2 at 0.7 V
- ANL has built a 1 inch diameter cell and obtained an open circuit voltage of 0.97 V

Development Issues for DMSOFCs

RESEARCH ISSUES:

Electrode materials development

Improvements in electrode polarization

Fabrication of thin electrolytes (2-20 μm) supported on an electrode

ENGINEERING ISSUES:

Thermal insulation and Packaging

Advantages of DMSOFC

A DMSOFC system offers many advantages over Polymer electrolyte fuel cell system operating on methanol:

Electrodes made of non noble metal materials

No methanol cross-over

Oxygen ion conducting electrolyte => no water management

Highly stable materials => no decomposition with temp. increase

Ambient pressure operation makes it simpler

Conclusions

Significant improvements must be made in electrode polarization to achieve high performance

Thin electrolyte fabrication needed to achieve high performance

A DMSOFC system offers many potential advantages over existing fuel cell technologies for portable power applications

A high performance DMSOFC system is potentially simpler than competing technologies

This is a promising technology that must be pursued further to realize its potential

**HEAT TRANSFER & COOLING CHALLENGES
IN FUEL CELL DEVELOPMENT**

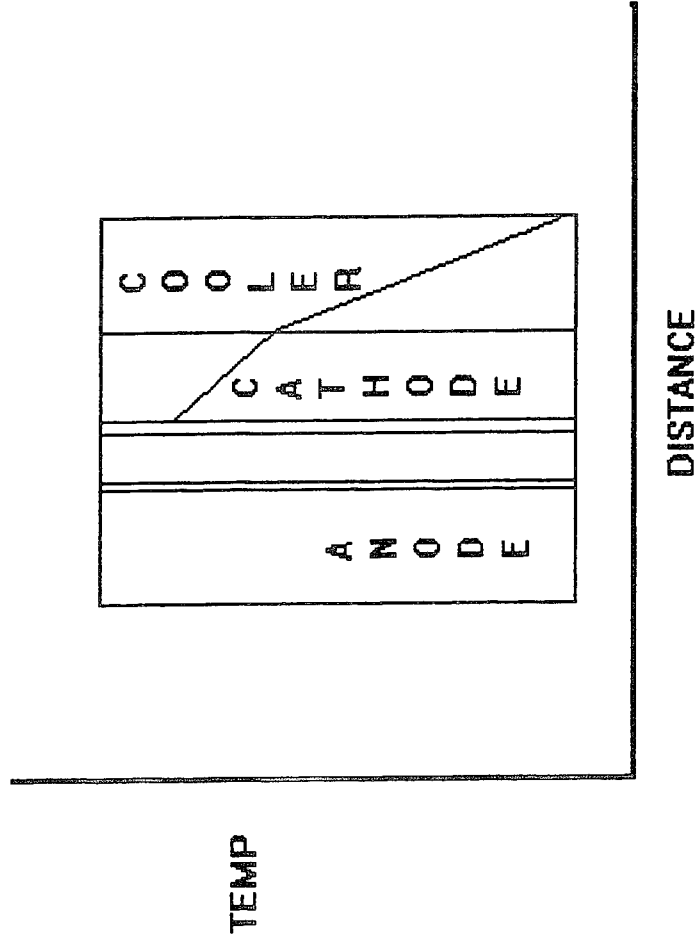
PROSPECTOR VII

OCTOBER 1994

**ANALYTIC POWER CORPORATION
PO BOX 1189
BOSTON, MA 02117**

**(617) 542-6352
(617) 695-3272 FAX**

AMBIENT PRESSURE PEM CELL
THERMAL MANAGMENT



ANALYTIC POWER CORPORATION

THERMAL MANAGEMENT IN AMBIENT PRESSURE PEM FUEL CELLS

ORIGINAL APPROACH

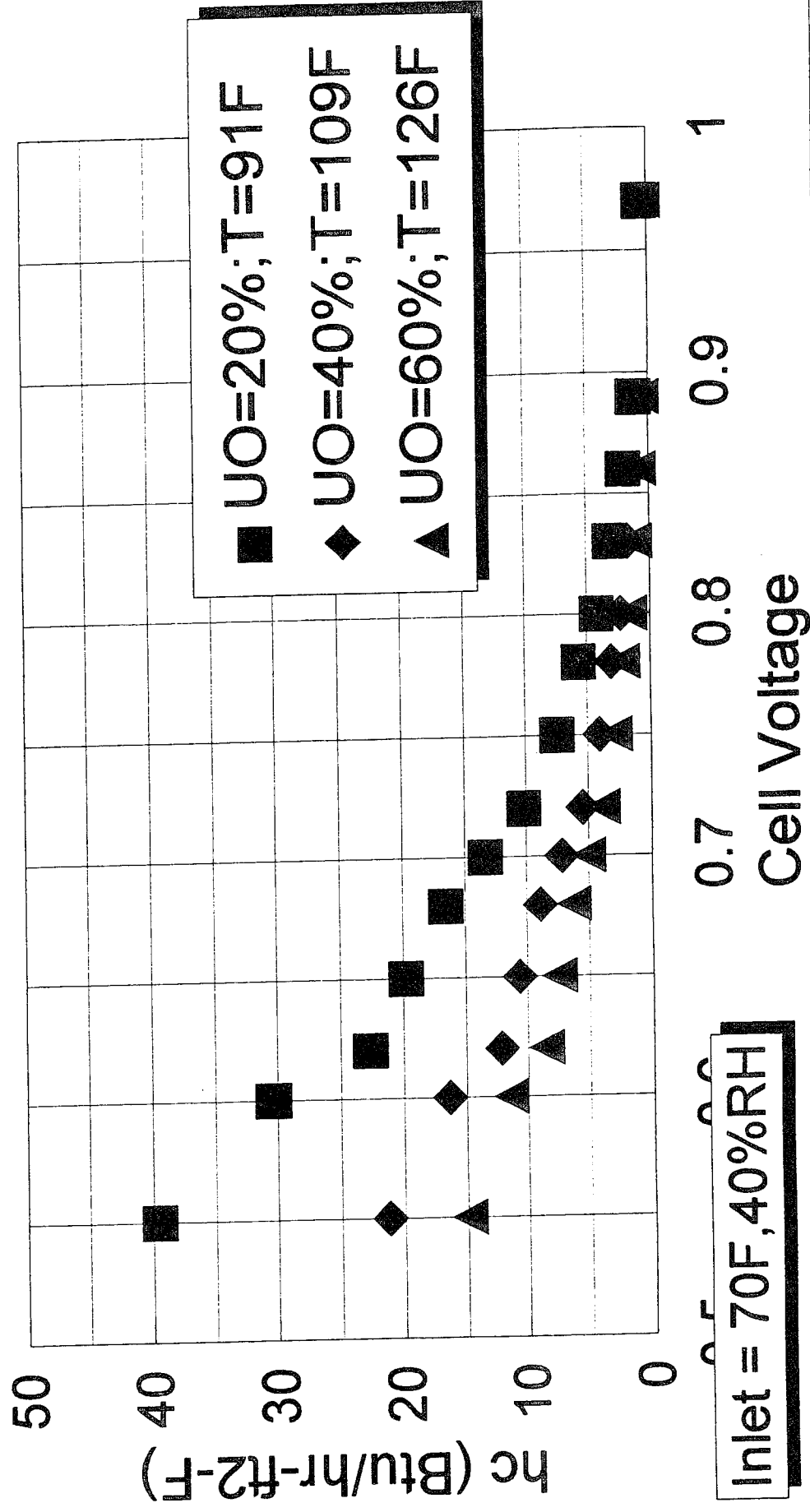
FREE CONVECTION - INTEGRAL COOLING AND REACTANT AIR

PROBLEM WITH APPROACH

- LOW OVERALL HEAT TRANSFER COEFFICIENT
- HIGH TEMPERATURE GRADIENT
- CURRENT DENSITY MALDISTRIBUTION
- POWER DOWN TRANSIENT

Min. Heat Transfer Coefficients

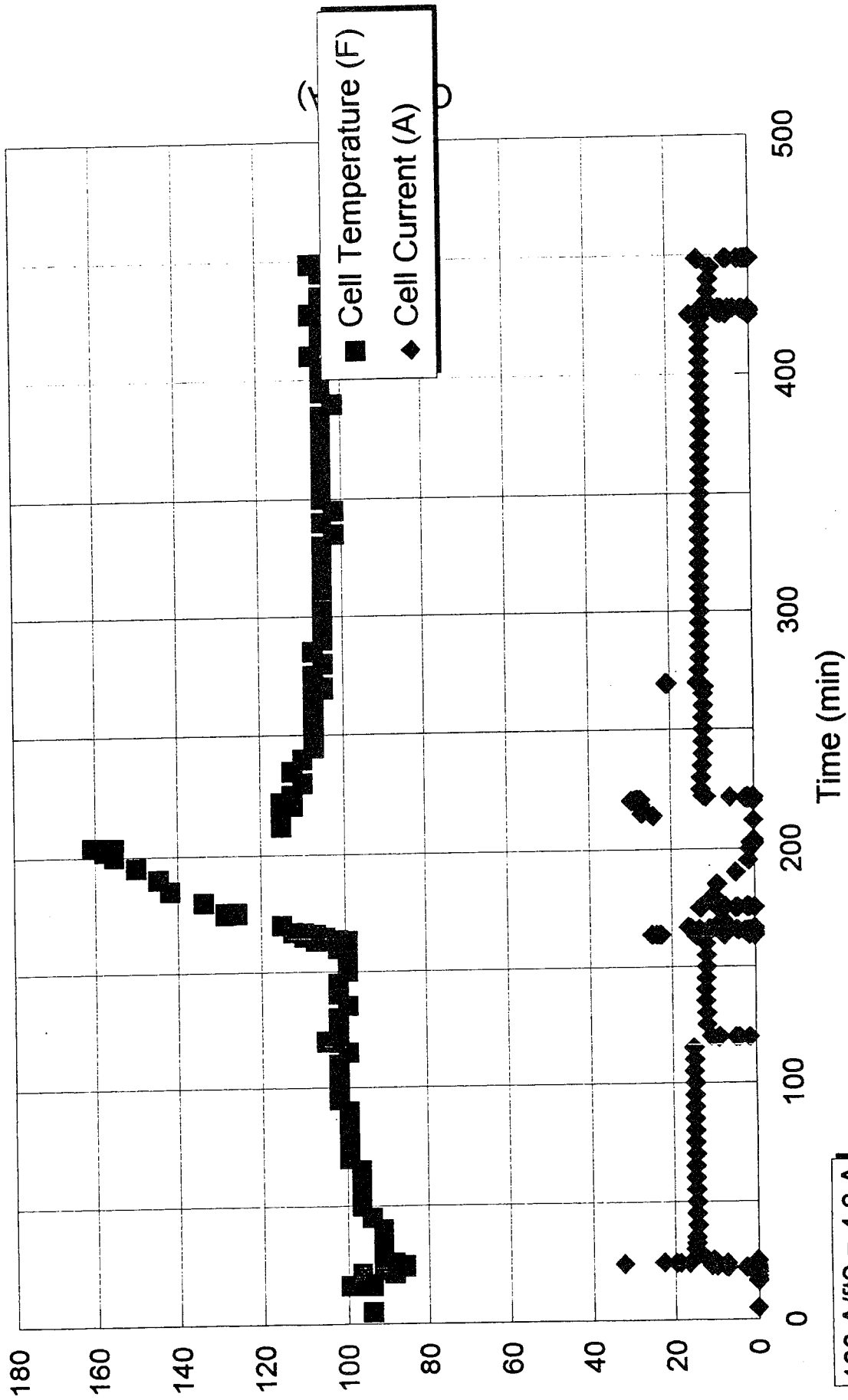
Equilibrium Cell Temps shown



Inlet = 70F, 40%RH

Cell Temperature & Current

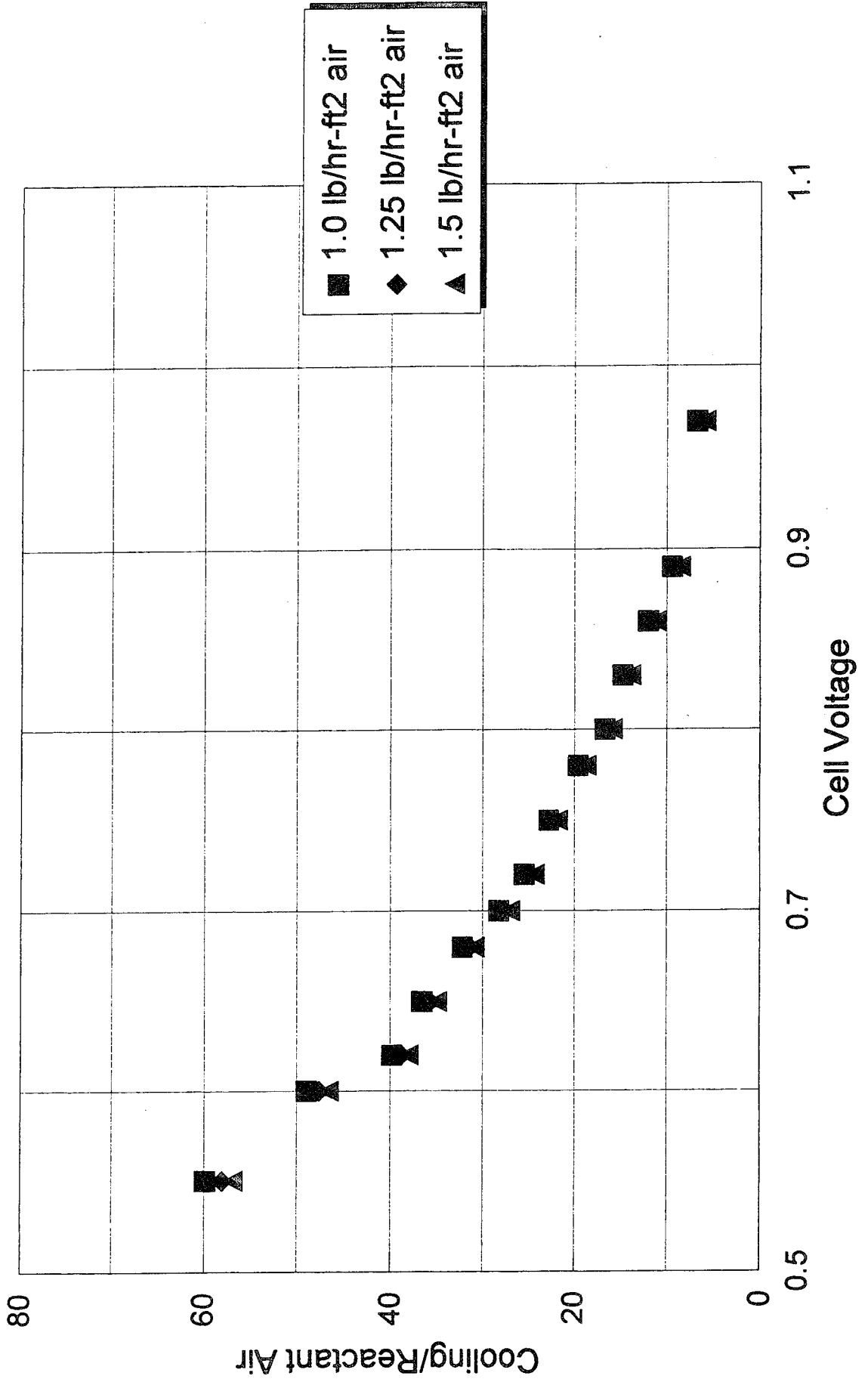
1 atm H₂/Air (dry)



100 A/ft² = 4.3 A

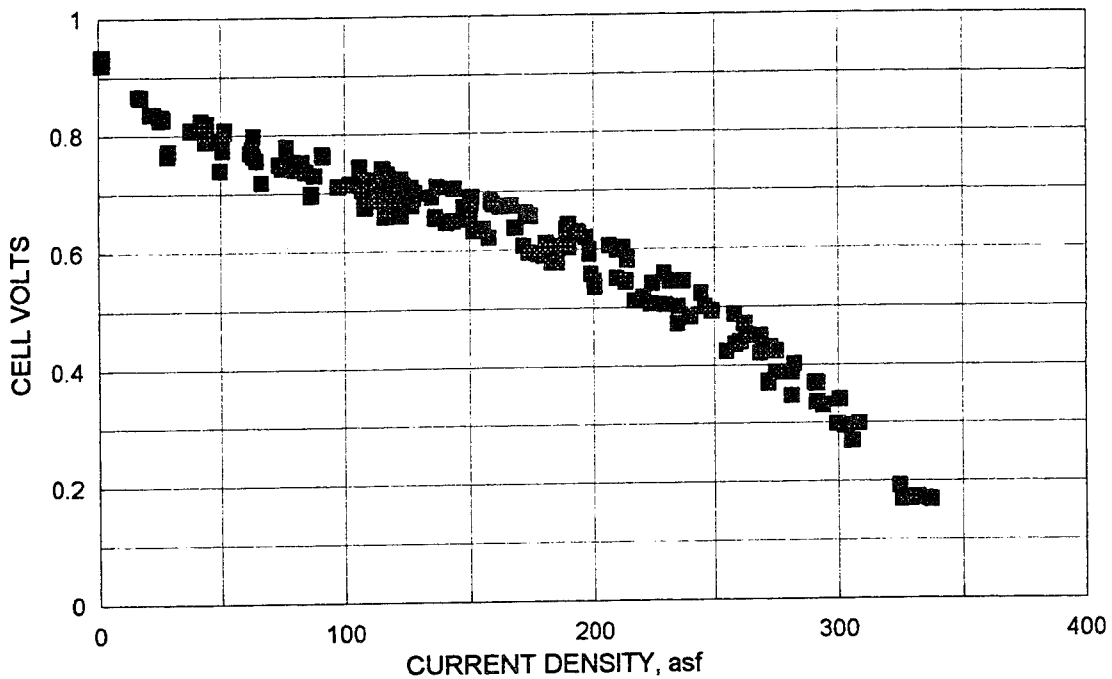
Cooling/Reactant Air Ratios

Constant Cathode Flows



VOLTS vs. CURRENT DENSITY

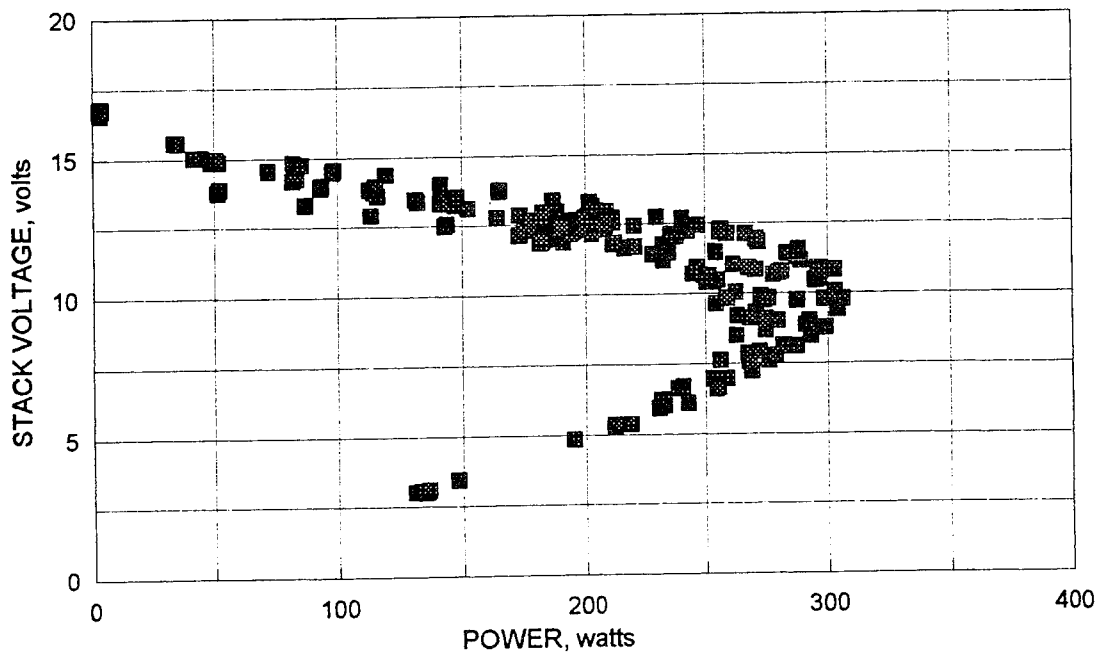
STACK#9 DATA E9011339.DAT, E9011418.DAT & E9011502.DAT



Niagara Mohawk 18 CELL STACK
H₂/AIR WITH 15V MULTIFAN REVERSE FLOW

STACK VOLTAGE vs. POWER

STACK#9 DATA E9011339.DAT, E9011418.DAT & E9011502.DAT



Niagara Mohawk 18 CELL STACK
H₂/AIR WITH 15V MULTIFAN REVERSE FLOW

CONCLUSIONS

- **FORCED CONVECTION - SEPARATE COOLING AND REACTANT AIR**
- **HIGHER OVERALL HEAT TRANSFER COEFFICIENTS**
- **ELIMINATES HIGH WATER LOSS AT HIGH COOLING AIR FLOW**
- **LOWER CELL TEMPERATURE**
- **LOWER TEMPERATURE GRADIENT**
- **MORE UNIFORM CURRENT DENSITY**
- **HIGH CELL PERFORMANCE OFFSETS INCREASED PART COUNT**

FIELD PRODUCTION OF HYDROGEN: WET OR DRY MANUFACTURE FROM MILITARY TRANSPORT FUELS

**William A. Peters
Associate Director
MIT Energy Laboratory**

Presented At

The PROSPECTOR VII WORKSHOP

**“Small Fuel Cells For Portable Power”
October 31 - November 3, 1994
Washington Duke Inn and Golf Club
Durham, NC**

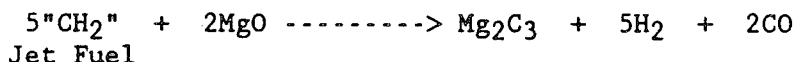
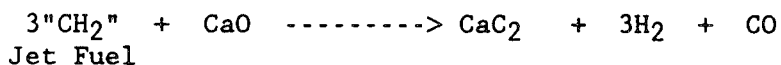
**Sponsored by Auburn University and
the U.S. Army Research Office**

NARRATIVE OVERVIEW

Two possible approaches for field manufacture of molecular hydrogen from logistic fuels like jet fuel or diesel, are considered in light of military needs for H₂ to power portable fuel cells.

In one approach, logistic fuel would undergo hydrolysis or partial oxidation¹ (with air) in supercritical water (SCW) (i.e. T > 374°C and P > 221 bar),² producing H₂, CO and probably other gases like methane. Water gas (WG) shift of the CO would double the H₂ yield, but would consume water e.g., up to one gal per gal of jet fuel converted. Sulfur in the logistic fuel could be separated from the SCW as inorganic sulfate salts by injecting caustic. Products yields and process efficiencies remain to be determined, but are expected to be adjustable by manipulating temperature, pressure, and other operating conditions. Maximum theoretical H₂ production is estimated at 160 standard cubic feet (SCF) per gallon of jet fuel converted (or 320 SCF with the WG shift). Without co-processing waste (see below) an additional 2/3 to 3/4 gal of jet fuel might be needed to supply process energy. In a 50% efficient fuel cell 160 SCF of H₂ will generate about 6.7 kWh of electricity. In practice, lower H₂ yields and significant production of methane or other hydrocarbons may arise because of equilibrium or kinetic limitations. Research is needed to shed light on the specifics. A process field-deployable in two ISO containers (8' wide x 8.5' high x 20' long) has the potential to supply one to two companies of soldiers each consuming 250 watt of electricity on a continuous basis (assuming a fuel cell efficiency of 50%). Further, the SCW process could operate on impure water and be used to destroy human or other toxic wastes. Such "dual use" applications could reduce or eliminate the need for other waste decontamination technologies, generate potable water, and provide a source of process heat for H₂ manufacture. Research is needed to determine yields of H₂ and other products from jet fuel and diesel fuel, desulfurization potential, and process thermal efficiency all as affected by practically relevant operating conditions e.g., temperature, pressure, feed concentration, feed/air ratio, and reaction time.

In the second approach, an electrical discharge (thermal plasma) reactor would be used to convert logistic fuel plus calcium oxide (or magnesium oxide or calcined dolomite) to H₂ according to the following global reactions:³



Theoretical maximum H₂ yields are about 160 SCF per gal of jet fuel converted. Fuel sulfur should be captured as CaS or MgS. Actual product yields, extents of sulfur recovery, and process thermal efficiency remain to be determined, but there is potential to achieve H₂ yields per kWh of electricity comparable to or better than the 7 SCF of H₂/kWh of conventional water electrolysis.⁴ However, because logistic fuel would be consumed as both the H₂ feedstock and (by combustion e.g., in diesel or gas turbine generators), to produce electricity to run the plasma, the total yield of H₂ per gallon of fuel consumed will be less, e.g., in the case of jet fuel perhaps 2/3 to 3/4 that of water electrolysis where the fuel is used only to

provide electricity. The plasma approach does generate valuable co-products like CaC_2 and, in contrast to electrolysis, does not consume water as its hydrogen source. More quantitative understanding of process fuel requirements and H_2 production potential requires research to determine yields of H_2 and co-products from logistic fuels, desulfurization capabilities, and process thermal efficiencies, as affected by operating conditions that can be manipulated in relatively portable, field-operable units, e.g., plasma power, CaO/fuel ratio.

If successful, both of these approaches would generate H_2 from diesel and jet fuel, using field-operable, portable equipment. Yields, thermal efficiencies, co-products, and effectiveness in sulfur capture remain to be established, but both methods have the potential to meet projected H_2 needs of forward operating military units on a continuous basis, using hardware that could be shipped in a small number of ISO containers and then field-assembled. Further, multiple military and civilian uses are possible with one or both of these technologies, e.g., wastes destruction, generation of other useful fuels like methane for field stoves, and production of potable water. Research along the lines discussed above is needed to assess the technical and economic feasibility of these two approaches and to provide data for comparing their performance potential with that of other technologies for field production of H_2 , e.g. electrolysis of water, high temperature reforming, and partial oxidation.

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3. Diaz, A.F., Modestino, A.J., Pride, J.D., Howard, J.B., Tester, J.W., Peters, W.A. "Conversion of Light Hydrocarbon Gases to Metal Carbides for Production of Liquid Fuels and Chemicals", Proceedings of the US DOE Coal Liquefaction and Gas Conversion Contractors' Review Conference, September 7-8, 1994, US DOE Pittsburgh Energy Technology Center, Pittsburgh, PA, in press (c. 1994).
4. Shreve, R.N., Brink, J.A., Jr. Chemical Process Industries, Fourth Edition, p. 99, McGraw-Hill, New York, (1977).

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PLEASE NOTE

**A Number of the Following Pages Have
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Presented at this Workshop**

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Workshop Hosts

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B. Phenix

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J. Pride

R. Lachance

J. Tester

P. Marrone

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ARO - URI

R. Shaw

DOE/PETC

**A. Bose
G. Cinquegrane**

PROPERTIES OF NORMAL AND SUPERCRITICAL WATER

	<u>25°C, 1 bar Normal Water</u>	<u>550°C, 250 bar SCW</u>
Density, g/cm³	1.0	0.1
Dielectric Constant, debyes	80	2
Ionic Dissociation Constant, (Mol/kg)²	10⁻¹⁴	10⁻²³
Viscosity, cp	1	0.03

A PRIMER ON SUPERCRITICAL WATER (SCW)

- **H₂O Above Its Critical Temperature (374°C) and Pressure (221 bar)**
 - **Physicochemically Different From "Normal" Water**
 - **An Unassociated, Non-Polar Solvent**
 - **O₂, N₂, CO, H₂ Totally Miscible**
 - **Many Organics Totally Miscible**
 - **Common Inorganics Virtually Insoluble**
 - **Oxidation of Organics Rapid and Thorough**
-

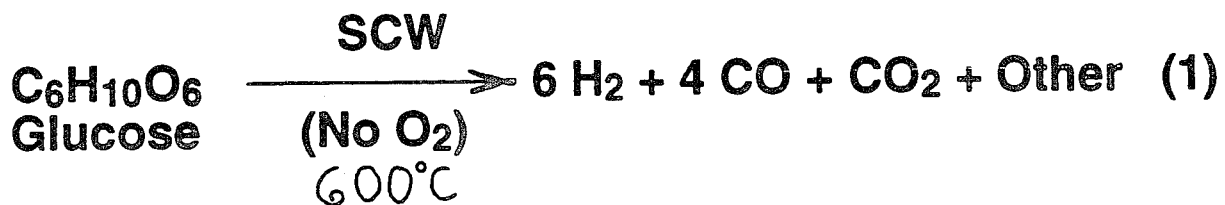
WET MANUFACTURE OF H₂

BACKGROUND

- **SCWO URI Kickoff Meeting: (Aug. 1992)**

R. Paor Raises Issue of H₂ From SCWO

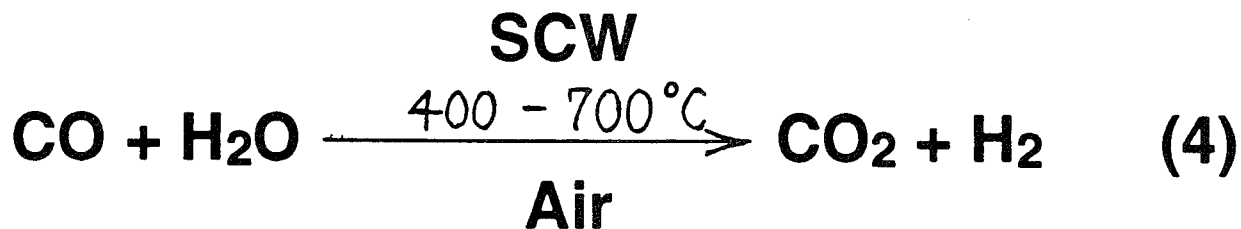
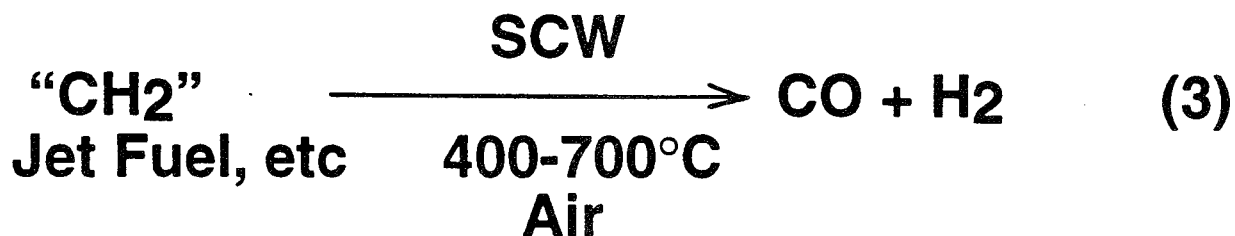
- **Holgate, Meyer, Tester (1992 +):**



WET MANUFACTURE OF H₂

PROPOSITION

Convert Jet and Other Military Fuels to H₂ By Oxidation in Supercritical Water:



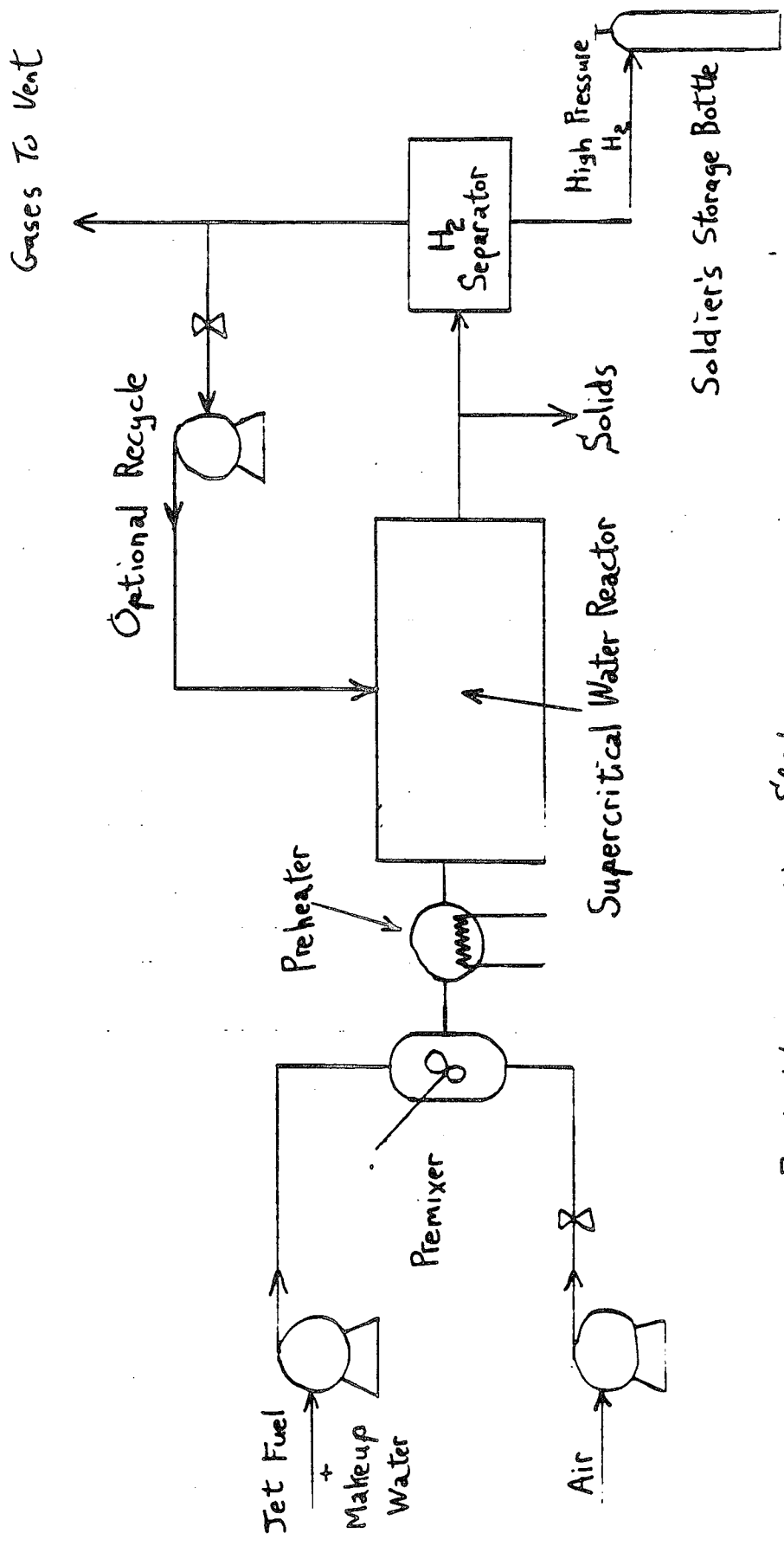
“POTENTIAL” (i.e. MAXIMUM THEORETICAL) YIELDS OF HYDROGEN BY REACTING JET FUEL IN SUPERCRITICAL WATER

	SCF H ₂	
	Per lb Jet Fuel ^a	Per Gal Jet Fuel ^a
No Water Gas (WG) Shift	25	160
Include WG Shift	50	320
Water Consumption in WG Shift, Gal	0.15	1

^aConverted, Process Heat May Require an Additional 2/3 to 3/4 Gallon Jet Fuel

DEFINITIONS AND CONVERSION FACTORS

- 1 “Power Warrior” (PW) = 1 Paur Warrior
 - 1 Power Warrior = 1 Soldier or Marine
Using 1/4 kW Continuously for 1 Day
= 6 kW-h \cong 0.9 lb H₂ (@ 50%)
 - 1 PW = H₂ From 1 Gal Jet Fuel (50%
Efficient Fuel Cell, No Water Gas Shift)
 - 1 PW = H₂ From 1/2 Gal Jet Fuel With WG
Shift BUT “Costs” 1/2 Gal of Water
 - 45 x 8 x 8.5 Feet High Trailor Truck
Mounted SCWO Process = 40 - 400 PW’s
(i.e. ~ 1 platoon to 2 companies)
-



Portable Mounting Skid

Figure 1. Sketch showing some major process elements in a portable system for field generation of hydrogen (H_2) by reacting jet fuel in supercritical water.

H₂ FROM SCWO OF JET FUEL, ETC. RESEARCH NEEDS

- **Technically Feasible For Jet Fuel, Diesel, etc.**
 - **Yields/Selectivity/Production Rates of H₂ For Practically Sensible Conditions**
 - **How to Turn Water Gas Shift On and Off**
 - **Other Feedstocks (Wastes)**
 - **Improve Yields, e.g. Supply 1000 PW's From One Trailer Truck**
 - **Compatibility With Dirty Water or Seawater**
 - **Corrosion**
 - **H₂ Separation & Purification**
 - **Economic Assessment**
-

H₂ BY SCWO OF JET FUEL, ETC., PROS AND CONS

- **Use Field-Available Feedstocks**
 - **H₂ Arrives Already Pressurized**
 - **Could Accommodate Impure Water**
 - **Field Manufacture Possible (Trailer Truck)**
-

- **High Pressure System**
 - **High Tech Chemical Processing**
 - **Would Like Higher Yields From Trailer Mounted Unit**
-

DRY MANUFACTURE OF H₂

A Primer on Plasmas and Chemical Processing

- A Plasma is an Ionized Gas
 - Degree of Ionization Can Vary Widely
 - Important Parameters
 - \vec{E}/P
 - Gas Type
 - \vec{H} (External)
-

THERMAL PLASMAS

$$T_g \cong T_e$$

- **Extremely High Temperatures**
 - **Gargantuan Heat Flux Densities**
 - **Expected Chemistries**
 - **Deep or Total "Reforming" of Feed to Constituent Elements, Ions & Excited States**
 - **Creation and Reaction of Energetic Intermediates**
 - **Survival of the "Chemically Fittest"**
 - **Rapid Quenching Very Important**
-

METHANE CONVERSION IN ELECTRICAL DISCHARGES

- ▶ **A Logical Way to Overcome Methane Stability**
- ▶ **Acetylene Manufactured by CH₄ Pyrolysis in Commercial Arcs**
 - **I.G. Farben (Huels, Germany)**
 - **DuPont (Montague, MI)**
- ▶ **Lower Yields and Selectivity**
- ▶ **Unwanted (Probably Toxic) By-Products**
 - **Soot**
 - **Tars**
 - **PAH**
- ▶ **Need for Rapid Quenching of Reactive Intermediates for:**

$$2 \text{CH}_4 \rightarrow \text{C}_2\text{H}_2 + 3 \text{H}_2$$
- ▶ **Without Adequate Quenching:**

$$(2 + X) \text{CH}_4 \rightarrow \text{C}_2\text{H}_2 + 3 \text{H}_2 + \text{tars} + \text{soot} + \text{etc.}$$

PROPOSED METHANE UPGRADING CHEMISTRY

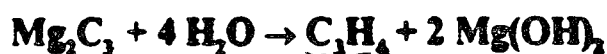
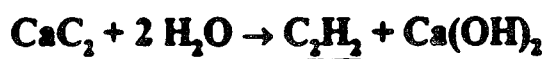
**Methane Reaction with Alkaline
Earth Metal Oxides:**



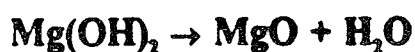
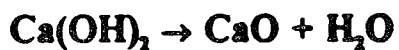
Chemical
Quench

**Separation, Storage and Transport of Solid
Metal Carbides (CaC₂ and Mg₂C₃)**

**Acetylene and Methylacetylene Production
from Carbide Hydrolysis:**



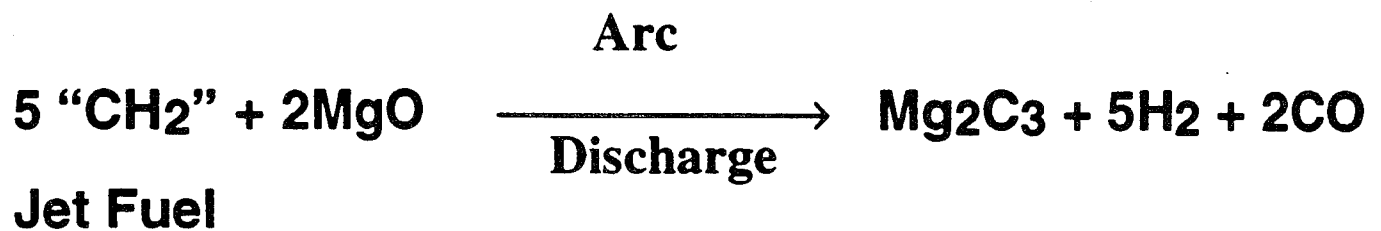
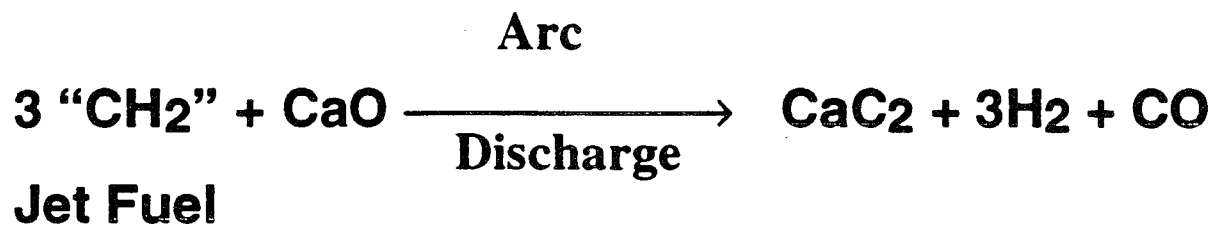
**Regeneration of CaO
and MgO for Recycle:**



**Oligomerization, etc. of
Acetylenes to Premium
Liquid Fuels and
Chemicals**

DRY MANUFACTURE OF H₂

Proposition



DRY MANUFACTURE OF H₂

Potential (i.e. Maximum Theoretical) Yields Per Gal. of Jet Fuel Converted^a

<u>Arc Discharge</u>	<u>CO (SCF)</u>	<u>H₂ (SCF)</u>	<u>H₂ (PW)</u>
CaO Route	53	160	1
MgO Route	64	160	1

^aAn additional 2 to 3 gal of jet fuel may be needed to supply electricity assuming electrical generator efficiencies of 25 to 30%. Water electrolysis to produce the same amount of H₂ would also require roughly this amount of jet fuel to generate the electricity.

DRY (ARC DISCHARGE) ROUTE TO H₂ - RESEARCH NEEDS

- **Technical Feasibility For Jet Fuel, Diesel, etc.**
 - **Thermal Efficiency**
 - **Yields/Selectivity/Production Rates of H₂ For Practically Sensible Operating Conditions**
 - **Hydrogen Separation & Purification**
 - **Economic Assessment**
 - **Scaleup of Plasma Reactor**
 - **Feasible w/o Plasma (Pure Thermal Routes)**
 - **Yields vs Portability**
 - **Management of CO & Carbides (Dual Use!)**
-

DRY (ARC DISCHARGE) ROUTE TO H₂ - BENEFITS & CHALLENGES

- **Field Compatible**
 - **Use Jet Fuel, Diesel, etc.**
 - **No Water Required**
 - **Potentially Significant Dual Use Pay Out
Since Acetylenes Are Major Co-Products**
-
- **Need Electricity**
 - **Low P So Need Chemical Storage or
Compression of H₂**
 - **High Tech Operation Especially to Make
Compact**
-

SUMMARY

- **Two Non-Conventional Routes to H₂ Proposed**
- **Both Would Use Standard Military Fuels As H₂ Source**
- **1 Gal Jet Fuel = 1 "Power Warrior"
≅ 0.9 lb H₂ ≅ 6.0 kW-h (@ 50%)**
- **Potential To Supply One to Two Companies of Soldiers From One Trailer Truck Mounted Processes**
- **Plasma Approach Requires No Water**
- **SCW Approach Could Operate In Limited Water Consumption Mode or Use Dirty Water**
- **Several Research and Development Challenges BUT**
- **High Potential Payout Including Significant Dual Use Potential**
 - **Wastes Destruction**
 - **Premium Co-Products**
 - **Potable Water**

UPDATE ON BATTERY WORKSHOP
Prospector III

Bob Guenther

Physics Division,
Army Research Office

The results of the Prospector III Workshop include new initiatives in power source systems designed for the individual soldier. The Soldier System has a 4500 W-hr mission requirement in the future. With that requirement, planners have examined the options available in the area of batteries, motor/generator systems, nuclear (radioisotope) source systems, and fuel cells. Only motor-generator and fuel cell power systems show near-term capability to reach the 4500 W-hr goal, thus Prospector VII is playing a vital role in shaping research and development program decisions dedicated to small, compact power sources for the SS. A table entitled "SS - 4500 W-hr Mission" shows the potential for improvement in the four candidate systems mentioned previously.

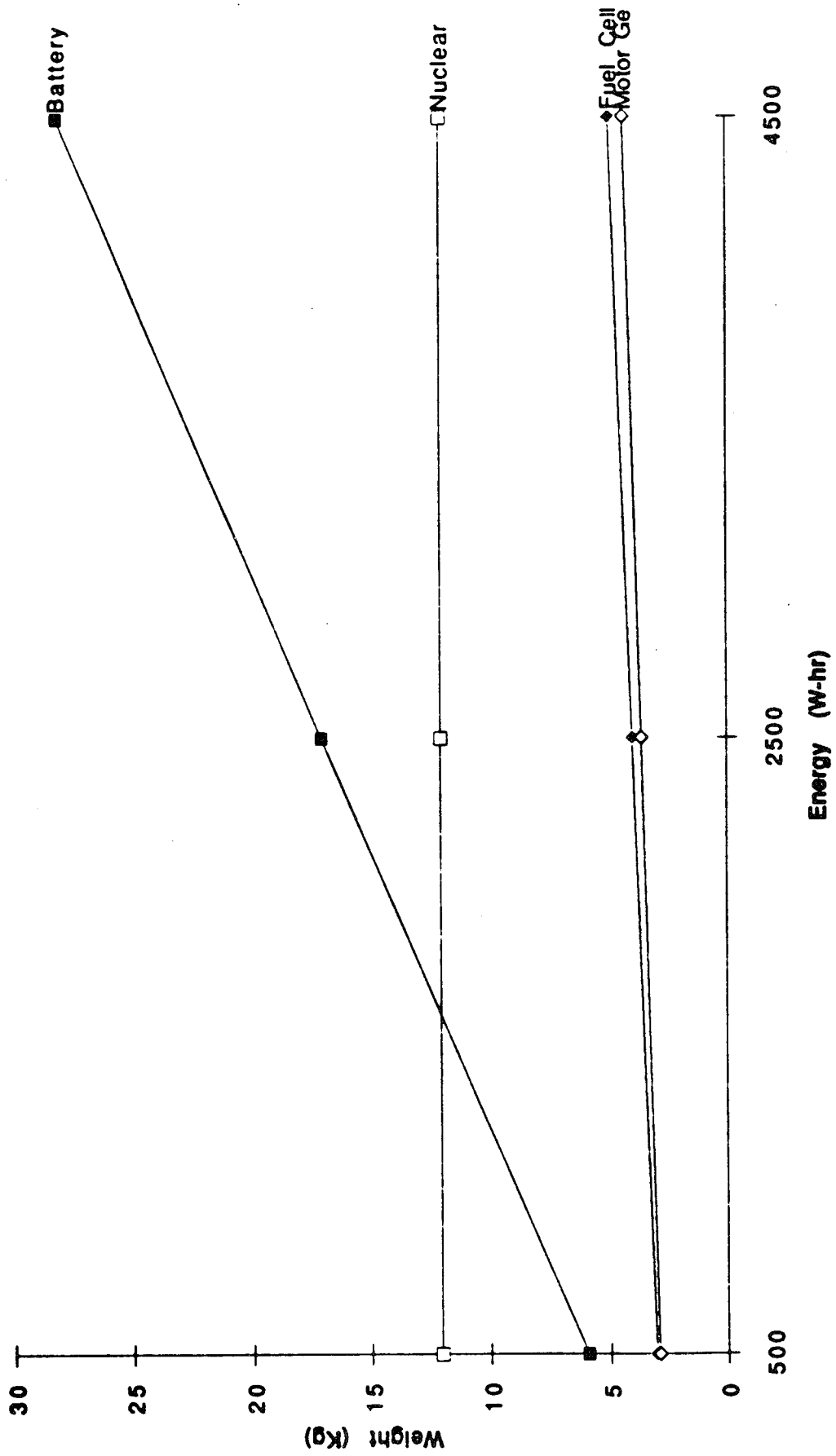
The 21st Century Land Warrior II System was outlined for Workshop participants. The major program components were discussed and related to supporting programs. System components were explained in terms of their weight and energy requirements.

Prospector VII
October 31 - November 3, 1994

UPDATE ON BATTERY WORKSHOP
(Prospector III)

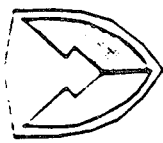
Bob Guenther
Director
Physics Division
Army Research Office

Weight to Energy Tradeoff Estimate



597

FAMILY OF STANDARD BATTERIES



ARMY
MATERIEL COMMAND



US ARMY
RESEARCH LABORATORY

CAPACITY CURRENT VOLTAGE	NON RECHARGEABLE LOW POWER Li/SO ₂	RECHARGEABLE LOW POWER Ni-Cd	NON RECHARGEABLE HIGH POWER Li/SO ₂	RECHARGEABLE HIGH POWER Ni-Cd
3	BA-5567/U			
6	BA-5372/U		BA-5847/U	BB-X847/U
12	BA-5588/U	BB-588/U		
24			BA-5590/U	BB-590/U

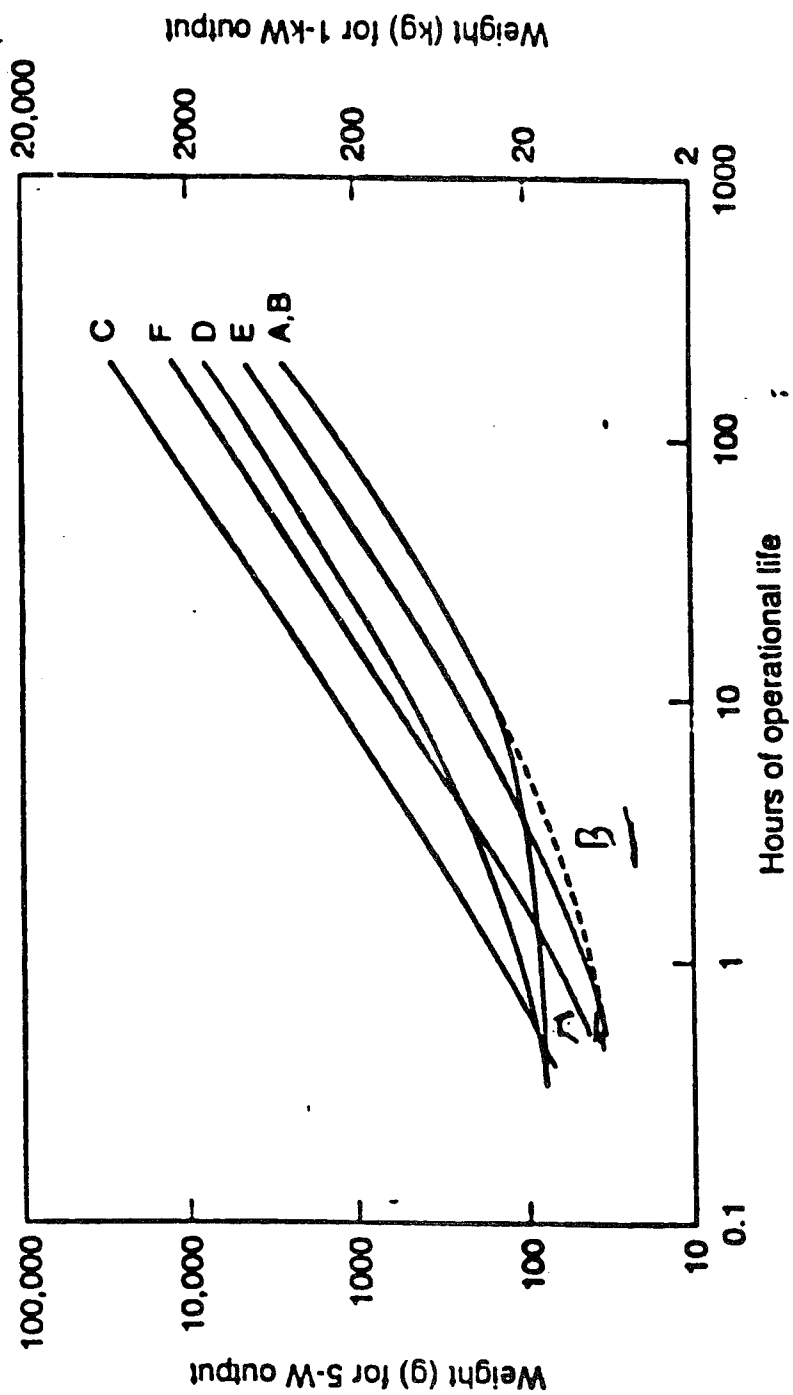


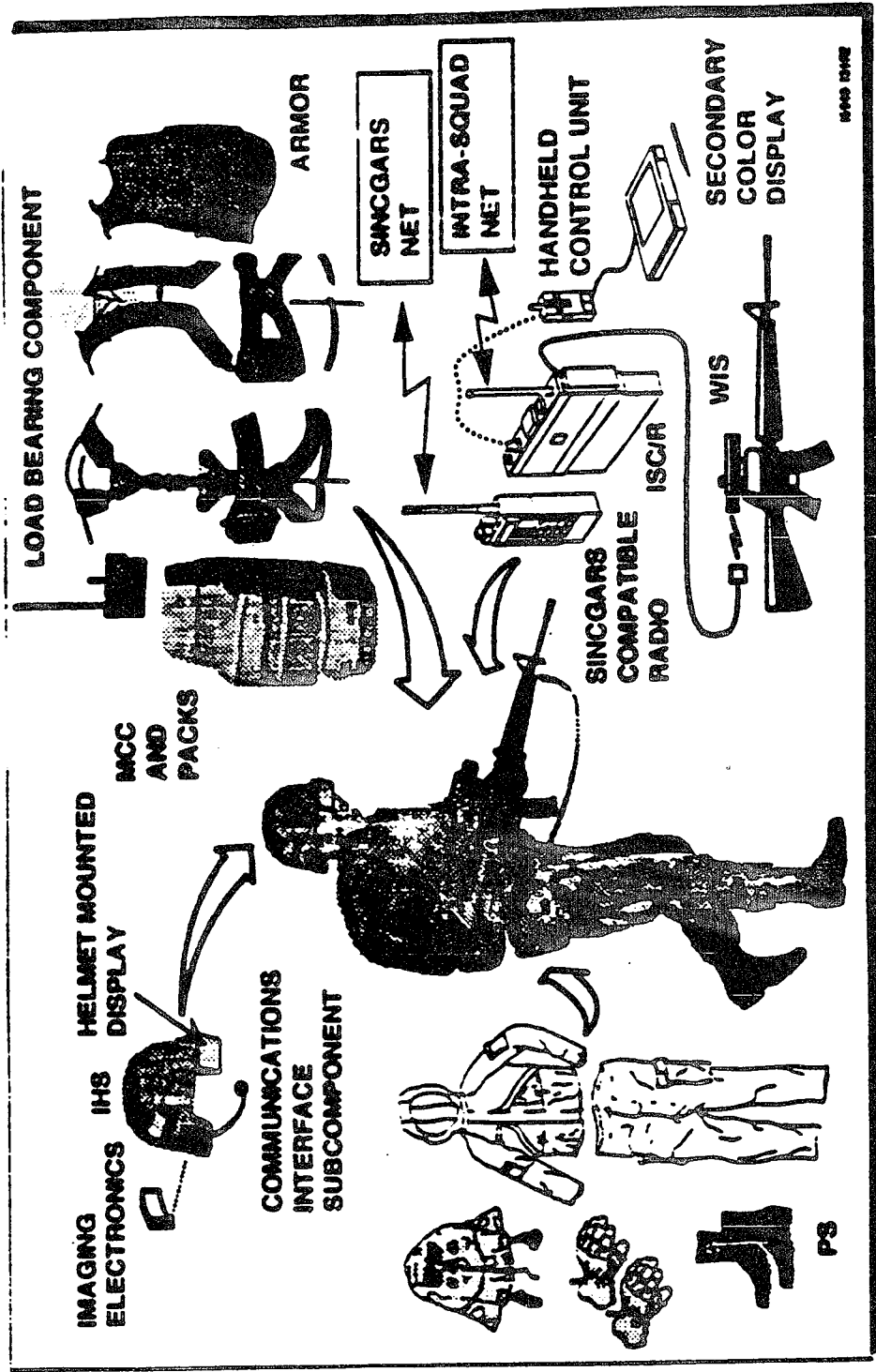
FIGURE 6.12 Comparison of electrochemical systems—weight vs. service life for 5-W and 1-kW output. A—Zn/air, 375 Wh/kg; B—Zn/air hybrid; C—Ni-Cd, 35 Wh/kg; D—Zn/MnO₂, 120 Wh/kg; E—Li/MnO₂, 225 Wh/kg; F—Li-ion, 85 Wh/kg.

(P)

6

21 CLW
 GEN II
 Soldier System

GEN II Soldier System



21st Century Land Warrior

Program Components

- **Gen II Soldier System ATD (System Contract)**
 - Integrated Headgear Subsystem
 - Individual Soldier Computer/Radio Subsystem
 - Weapons Interface Subsystem
 - Protective Subsystem
 - Microclimate Cooling Subsystem (battery-powered)
- **Supporting Programs**
 - Objective Individual Combat Weapon (OICW) TD
 - Integrated Sight Modules (ISM) TD
 - Advanced Image Intensifier (AI2)
 - High Resolution Flat Panel Display System TD
 - Multi-Purpose Individual Munition (MPIM) TD
 - FO/FAC ATD (USMC)
 - Individual Mine Detection TD
 - Combat ID TD
 - Individual Soldier Power TD

21 CLW

GEN II

Soldier System

System Components

- **Integrated Headgear Subsystem (IHS)**
 - Composed of Helmet Mounted Display, Head Mounted Mobility Sensor, Ballistic Shell and Communications Interface
 - Maximum head borne weight is 5.2 lbs.
- **Individual Soldier's Computer/Radio (ISC/R)**
 - Computer processes C2 and situational awareness software
 - Inter and Intrasquad radios link soldier to the digitized battlefield
 - GPS, navigation, voice recognition, image processing and other system interfaces
 - 2-4 lbs weigh requirement, 12 hour continuous operation
- **Weapon Interface Subsystem (WIS)**
 - Interface the ISC/R and IHS to the TWS and OICW
- **Protective Subsystem (PS)**
- **MicroClimate Cooling Subsystem (MCC)**
 - Provides 4 hours of metabolic cooling
 - 10 lb maximum weight including power source



MOTOROLA

**DEMONSTRATIONS, DISPLAYS
&
POSTER SESSIONS**

DEMONSTRATION, DISPLAY & POSTER SESSION

Chairman: Dr. Richard Paur (ARO)

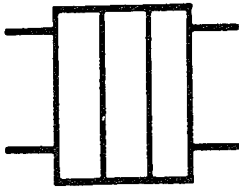
Four events, a demonstration, display, and poster session of portable fuel cell technology, provided enrichment to the technical presentations and "hands on" opportunities for participants.

- A. Demonstration/Display: Analytic Power's Operational Fuel Cell (Bloomfield)
- B. Display: CECOM's Fuel Cell Development Device (Taschek)
- C. Poster Session: Factors Governing On-Line Hydrogen Generation from Aluminum in Alkaline Solution (Rao)
- D. Poster Session: PEM Fuel Cells - Basic Research to Tech Development (Srinivasan)

The fifty-three participants, drawn from industry (19), academia (12), and government agencies and laboratories (22), represent a balanced cross-section of fuel cell researchers, users, and technologists.

The remainder of this document is a collection of the Workshop presentations and summaries from the individual Working Groups. The Appendices contain additional documentation or background papers as necessary to provide a complete written record of the events of Prospector VII.

POSTER SESSION A



ANALYTIC POWER CORPORATION

(617) 542-6352, FAX (617) 695-3272, BOX 1189, BOSTON, MA 02117

INTRODUCTION TO ANALYTIC POWER

Analytic Power, a privately held Massachusetts Corporation since 1984, has expertise in mechanical, chemical and electrochemical engineering. We have won nine Phase I and four Phase II Small Business Innovative Research (SBIR) awards, an Army SBIR Phase II Quality Award, and U.S. Patents for Unitized Fuel Cells and Electrochemical Refrigeration. We work for private industry and the government.

We do research and development in fuel cells, batteries, compressors, thermal systems and fuel processing systems. Our commercial line includes the recently announced Century Series Fuel Cell Power Supplies, a portable chemical hydride hydrogen generator and hydrocarbon reforming hydrogen generators.

Our Phase II SBIR program with the Army Research Office yielded Analytic Power's 25 Watt to 500 Watt fuel cell power supplies. These compact, air breathing power supplies, which can be equipped with a very compact hydrogen source, are suitable for applications requiring very high energy densities. This program won an Army SBIR Phase II Quality Award.

A Navy Phase II SBIR program is sponsoring a 10 KW fuel cell power plant for naval surface vessels. Our development program includes a high performance membrane fuel cell stack, an autothermal reformer based diesel fuel processor, a water and thermal management subsystem, a turbocharger based air processing system, and an integrated control system.

Microclimate cooling is the focus of our electrochemically driven heat pump program with the Navy Clothing and Textile Research Center. In earlier electrochemical compressor work we built an electropneumatic robot actuator with assistance from MIT's Robotics Lab and a grant from the National Science Foundation. The linear motor free piston compressor we are constructing with ARPA support will bring the advantages of pressurization to miniature power supplies. We are investigating a novel, planar, low temperature (650 C) solid oxide fuel cell using fabrication methods common to the electronics industry. This work is being done with cooperation from Tufts University and support from an NSF Phase I SBIR.

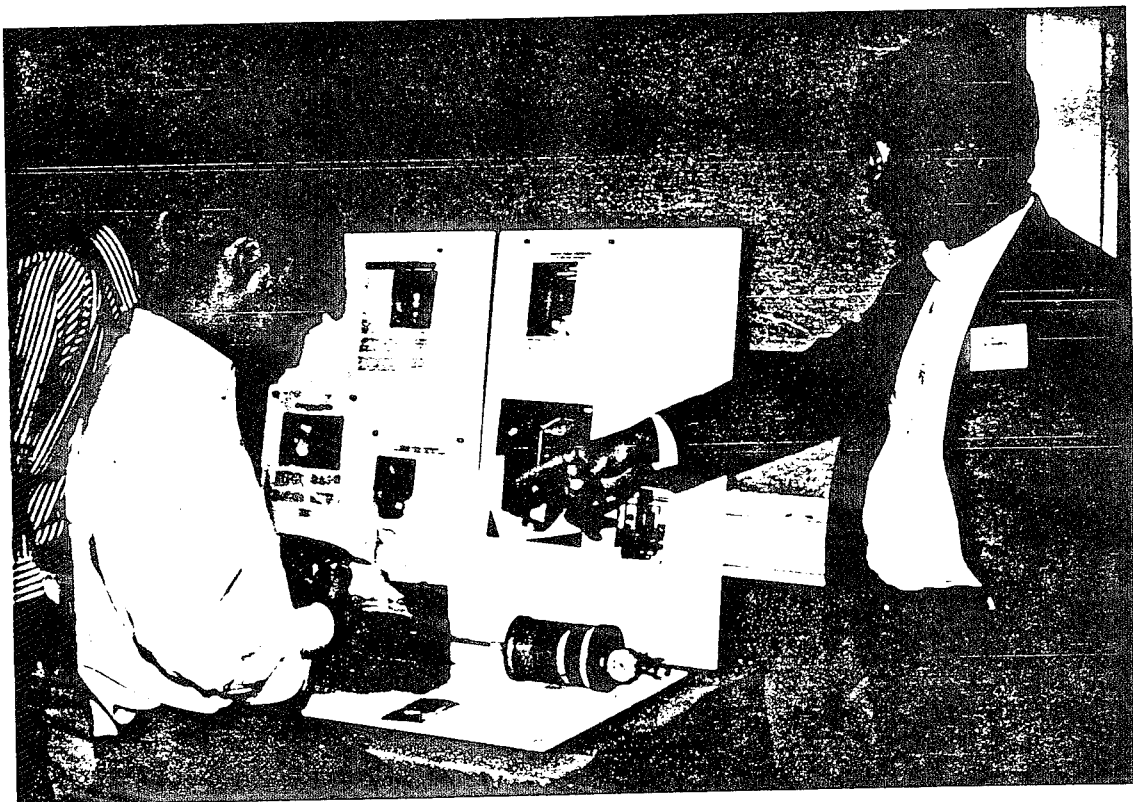
Systems analysis is used in all of our internal programs and we conduct systems analysis studies for others. The Naval Surface Warfare Center used Analytic Power's systems analysis programs to assess the impact of fuel cell technology on the design, cost and effectiveness of surface combatants as published in "The Assessment of Fuel Cell Power Plants for Surface Combatants."

Our 6100 ft² R&D facility is located in Boston, Mass.. Our laboratory includes ten stations for testing single cells, fuel cell substacks and full stacks, heat exchangers, linear motor free piston compressors, oxygen and hydrogen electrochemical compressors, and integrated fuel cell power plants. All of our test stands operate with in-house developed data acquisition and control.

Analytic Power's prototype factory has a manufacturing capacity of sixteen fuel cell stacks per month. The factory includes hydrolysis facilities, a carbon milling machine, and electrolytic treatment and vacuum casting facilities. All of these were designed and built by Analytic Power. We also have a commercial screen printer, presses with customized temperature control systems, and several ovens.

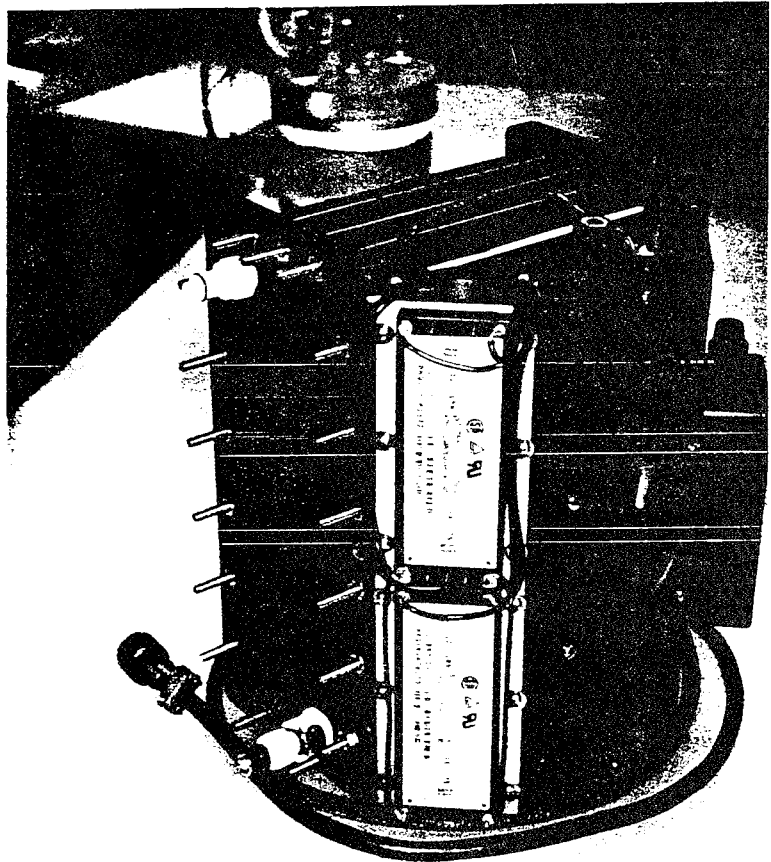
Analytic Power has established working relationships with vendors in many disciplines including thermoplastic injection molding, adhesives, machining, catalysts for fuel cell and fuel processing, and carbon composite structures. We have an extensive collection of literature in chemistry, electrochemistry, chemical engineering, mechanical engineering, materials and computer sciences. We are conveniently located near several large libraries including MIT, Harvard, Hanscom Field and the Boston Public Library. Our location gives us quick access to energy and engineering information.

Analytic Power can handle every phase of energy related research and development



ANALYTIC POWER CORPORATION CENTURY SERIES FUEL CELL POWER SUPPLIES

Analytic Power announces the Century Series Fuel Cell Power Supplies. These power supplies operate on hydrogen and air and produce electricity and water. The hydrogen can be supplied from regulated gas bottles or it can be generated by Analytic Power's Fuel Pacs. The power supplies can be operated between 40°F and 120°F. The natural voltage regulation is about 37% from full to no load. Analytic Power can supply DC to DC voltage regulators if required.



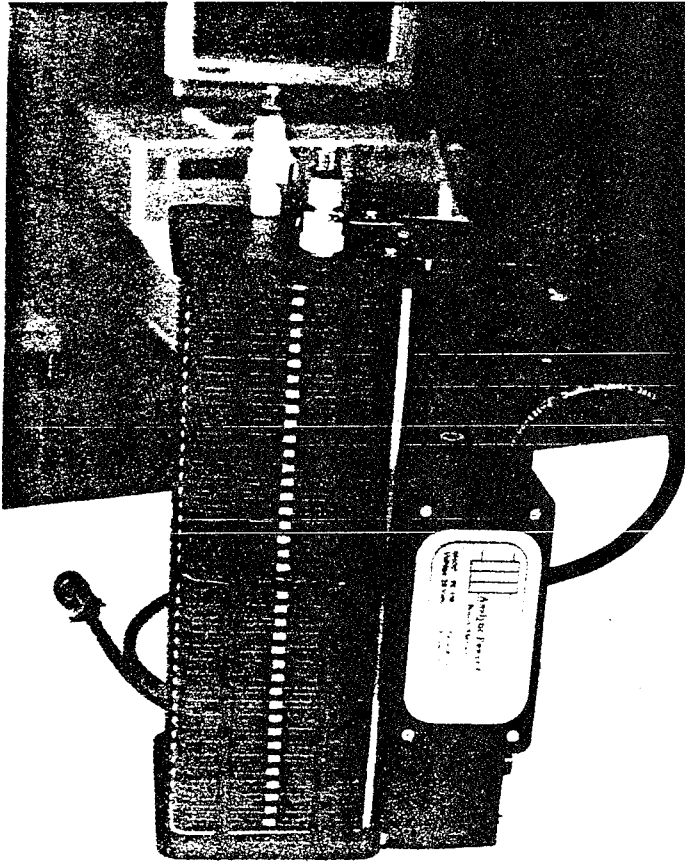
Analytic Power FC-200 Fuel Cell Power Supply

In fuel cells, unlike batteries, the fuel supply and the electrical generator are separate. The fuel cell is similar to an engine and generator set except that the efficiency can range between 56% and 77%. The chemical hydride Fuel Pacs can give the fuel cell energy densities of 800 to 1,400 watt hours/pound.

Based on the same technology used for powering space craft, the fuel cell is silent and pollution free. The Prototype FC-200 in the photograph was developed by Analytic Power for the Army Research Office. It recently powered a three-wheeled scooter and replaced lithium batteries to operate a microclimate cooler.

ANALYTIC POWER CORPORATION

FC-150 SMALL FORMAT FUEL CELL STACK



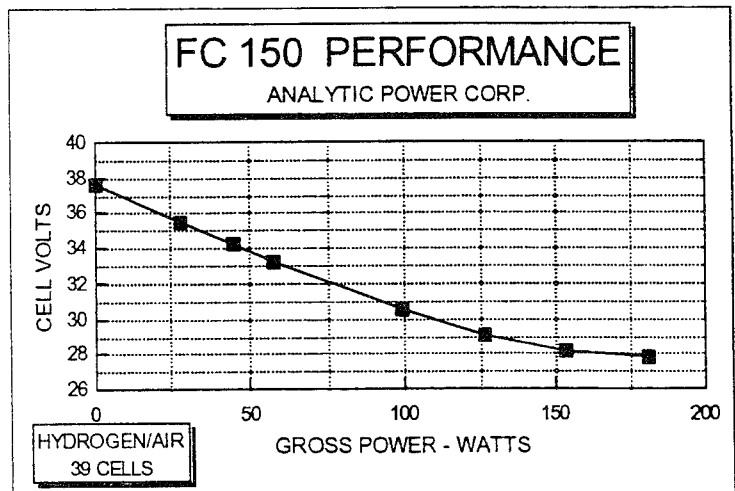
Analytic Power announces the FC-150, the most recent addition to our Century Series Power Plants. The FC-150, like other members of the series, operates on air and hydrogen supplied either from gas bottles or Analytic Power's Fuel Pacs. The 39 cell stack develops 28 Volts at a power level of only 150 Watts. The small format fuel cells are a mere 2.5 inches square.

Temperature Range: 40°F - 120°F
Voltage Range: 28V (load) - 38V (open)
Efficiency: 56% - 77%
Voltage Regulation: Natural

Fuel cells provide silent, pollution free power. The FC-150 is being built now. Its large format relatives, the FC-100 and FC-200, have been operating for months at rated power.

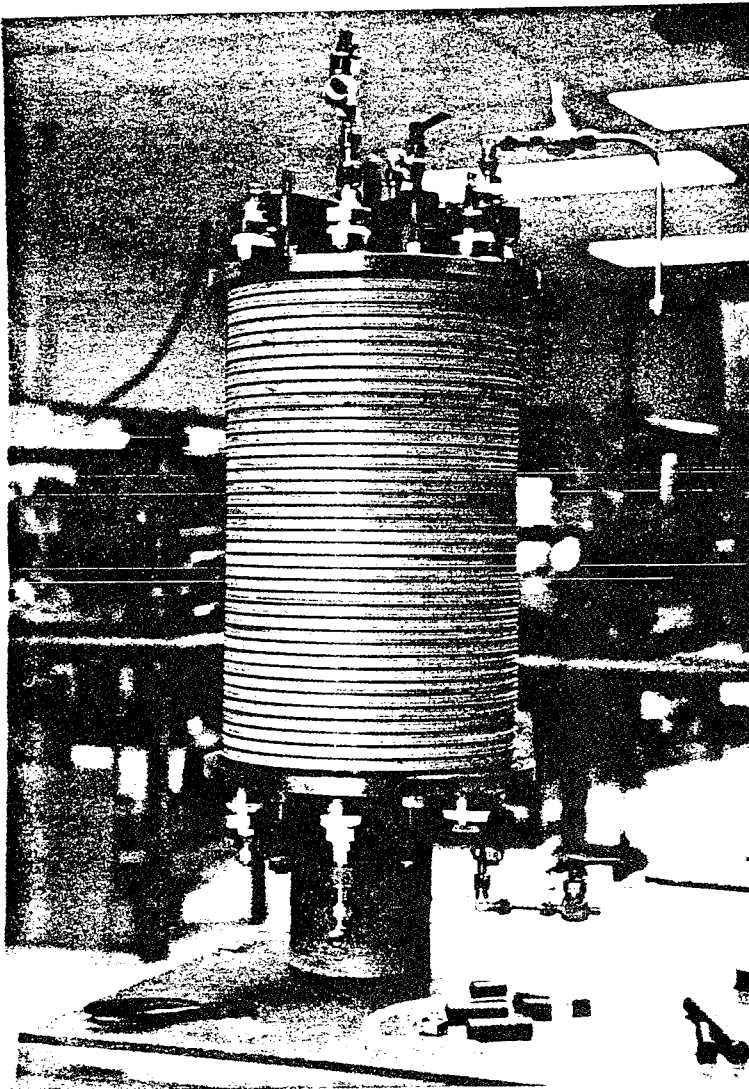
FC-150 Specification

Power	153 watts
Voltage	28 volts
Current	5 amp
Cell Voltage	0.723 volts
Current Density	125 ASF
Number of Cells	39
Length	11.5 in.
Width	4 in.
Height	5.75 in.
Weight	8.1 lb.
Volume	265 in. ³
Estimated Cost	\$8,650



ANALYTIC POWER CORPORATION

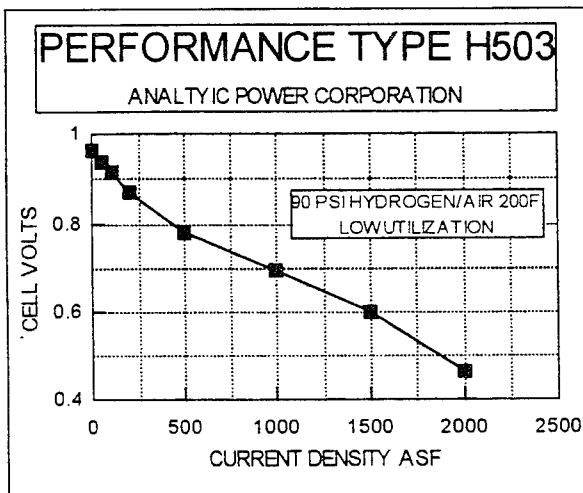
FC-10K Fuel Cell Stack



Analytic Power announces the FC-10K, a ten Kilowatt Fuel Cell Stack. The FC-10K operates on hydrogen or reformed hydrocarbon fuel and either pressurized air or oxygen. The fuel cell stack and a hydrocarbon fuel processor are being developed in an Office of Naval Technology SBIR program as a demonstrator for surface ship auxiliary and propulsion power. The FC-10K is also suitable for undersea applications.

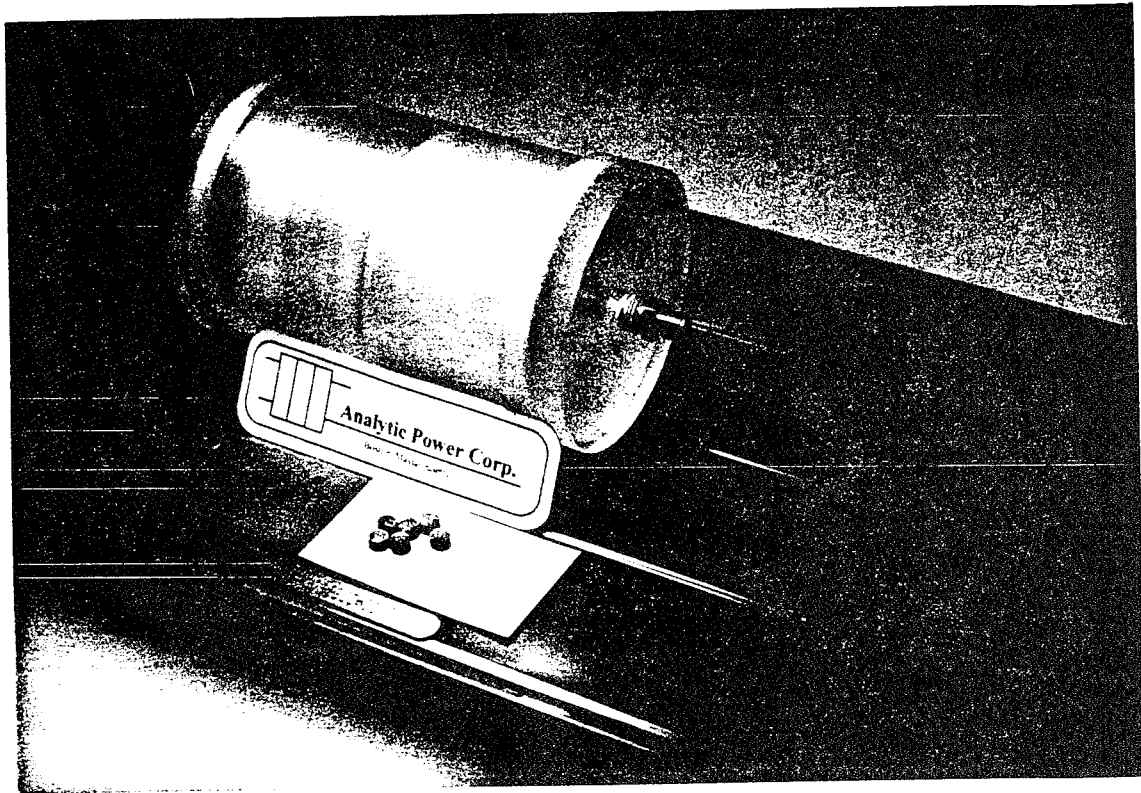
The water cooled FC-10K is a stack of 9.8 inch diameter circular "twin packs" separated by intercell coolers with advanced titanium bipolar plates. Sixty cells and thirty coolers make up a 10.7 inch high stack.

Like all fuel cells, the FC-10K is a silent, pollution free, power generator. The fuel cell efficiency of 56% at its rated power point increases as load is reduced. The FC-10K is in development today. Analytic Power's Century Series fuel cell power supplies, the FC100 and FC200, have operated for months at rated power.



<i>Temperature Range:</i>	120°F to 180°F
<i>Voltage Range:</i>	42V (load) - 58V (open)
<i>Voltage Regulation:</i>	Natural
<i>Pressure:</i>	150 psig maximum
<i>Power:</i>	10.3 kW
<i>Current Density:</i>	1000 ASF
<i>Weight:</i>	48 Lbs
<i>Volume:</i>	265 in ³

ANALYTIC POWER CORPORATION FUEL PAC 750



Analytic Power Corporation announces the Fuel Pac-750. The Fuel Pac-750 will produce 15.5 cubic feet (440 liters) of hydrogen, enough for 750 watt hours of energy with Analytic Power's Century Series Fuel Cells.

The Fuel Pac produces hydrogen on demand by reacting the chemical hydride fuel cartridge with the water stored in the Fuel Pac. The Fuel Pac can be started and

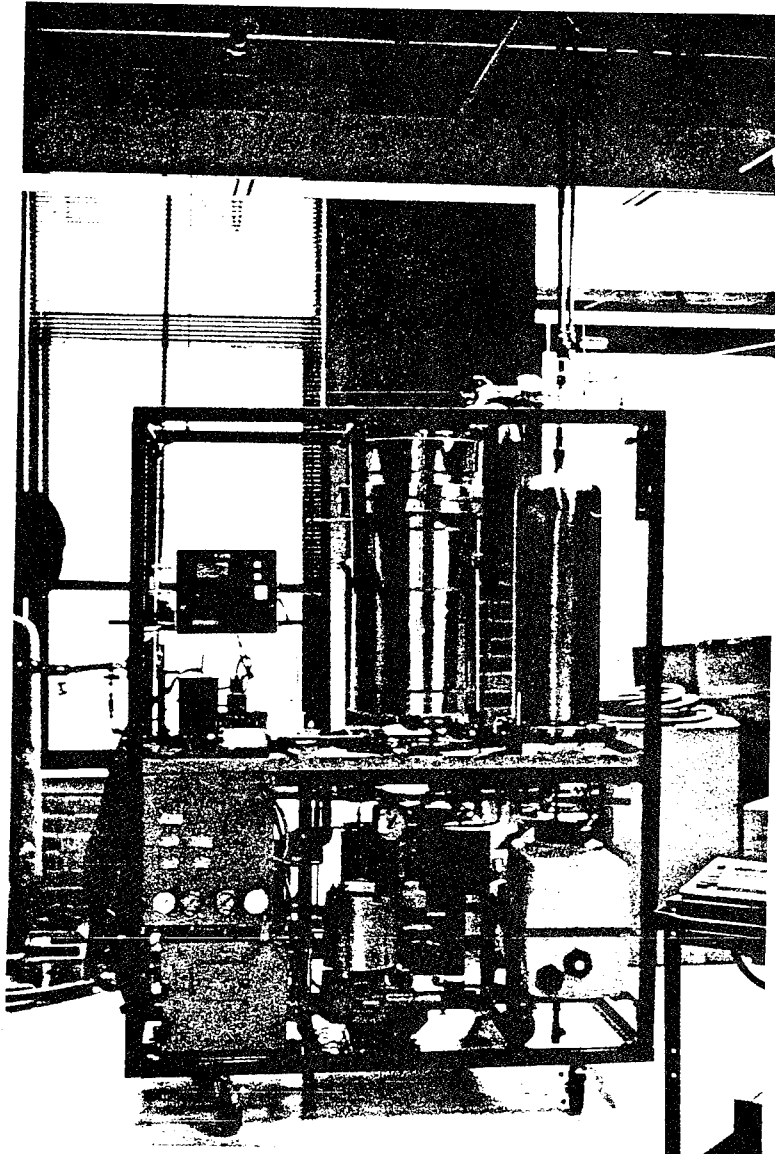
stopped. It is position insensitive, operating equally well on its side or inverted. The Fuel Pac fuel cartridge is available from Analytic Power.

Fuel Pac-750 Specification

<i>Weight:</i>	<i>3.5 pounds</i>
<i>Dimensions:</i>	<i>4.5" dia. x 6.5" long</i>
<i>Cartridge Weight:</i>	<i>80 grams</i>

*Analytic Power Corp.
PO Box 1189
Boston, MA 02117*

ANALYTIC POWER CORPORATION HYDROCARBON FUEL PROCESSOR



Analytic Power Corporation announces the Hydrocarbon Fuel Processor (HFP). The HFP produces 4.6 cfm of hydrogen from 4.7 pounds of sulfur bearing (0.25%) diesel fuel per hour. All lower molecular weight hydrocarbons including kerosene, propane and natural gas can be processed

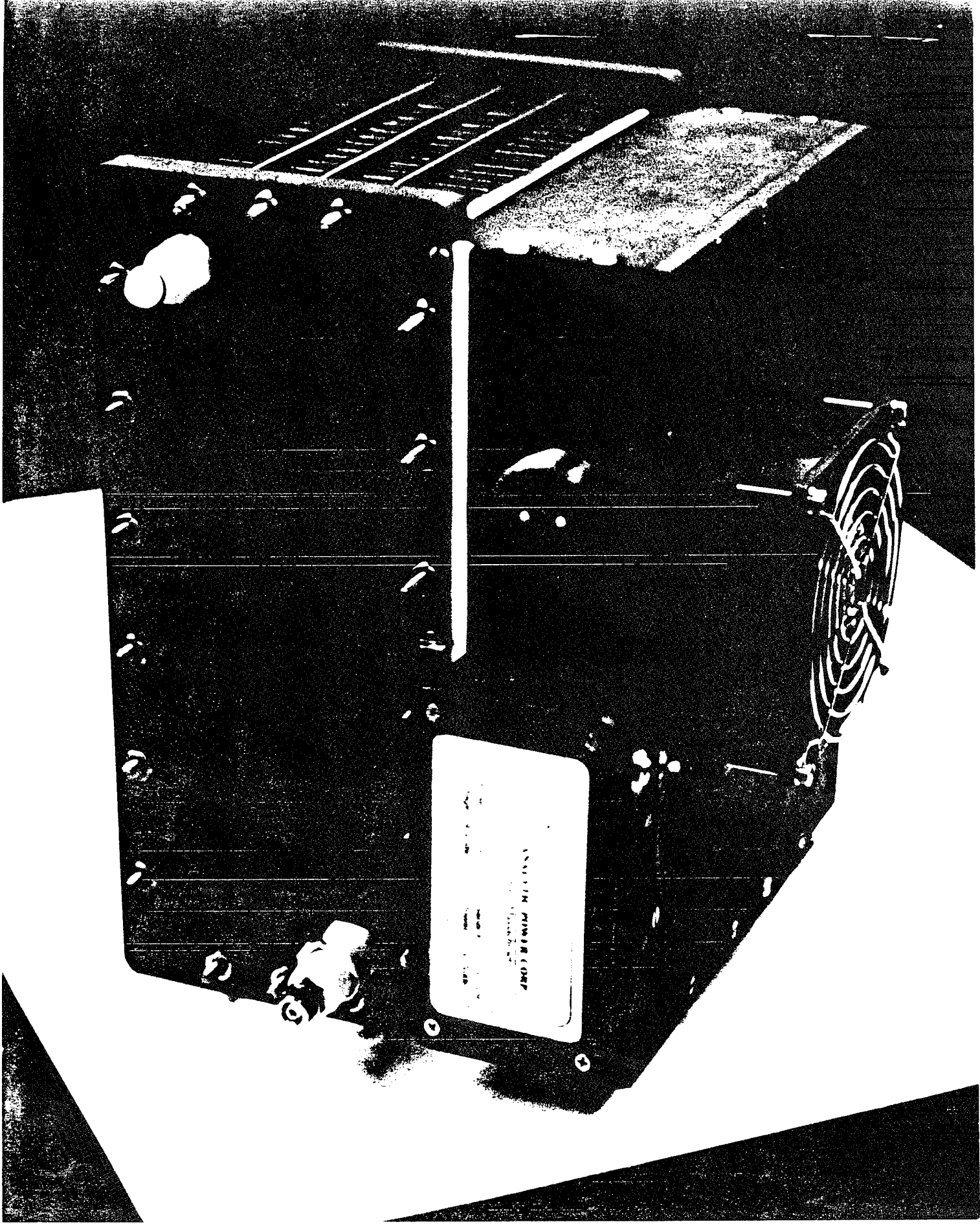
Designed for stand-alone operation, the HFP includes an auto-thermal reformer, shift converter, sulfur scrubber, and a package boiler.

The HFP is fully instrumented including an IR spectrometer for measuring carbon dioxide levels in the reformer effluent.

SPECIFICATION	
HEIGHT	5.4 FEET
WIDTH	5.0 FEET
DEPTH	2.5 FEET
WEIGHT	1200 LBS [†]
POWER	50 AMP, 3-PHASE, 220 VOLTS
PRICE	\$50,000 [‡]

[†] With full tanks
[‡] FOB Boston including operator's manual and simulation software

*Analytic Power Corp.
PO Box 1189
Boston, MA 02117
617-542-6352*

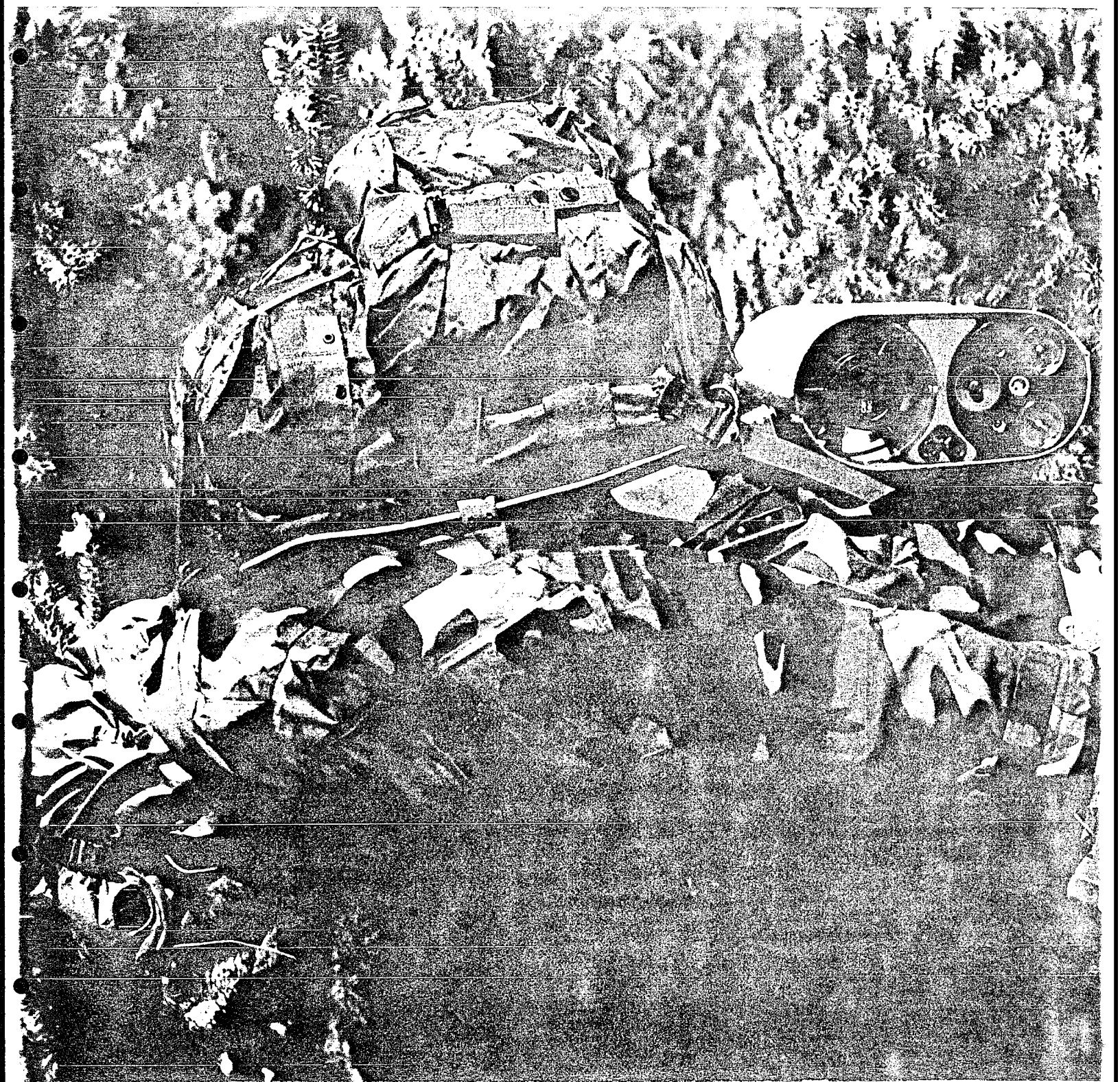


POSTER SESSION B

**CONCEPT FOR
FUEL CELL
POWERED
COOLING
MICROCLIMATE
BACKPACK
MODULE**

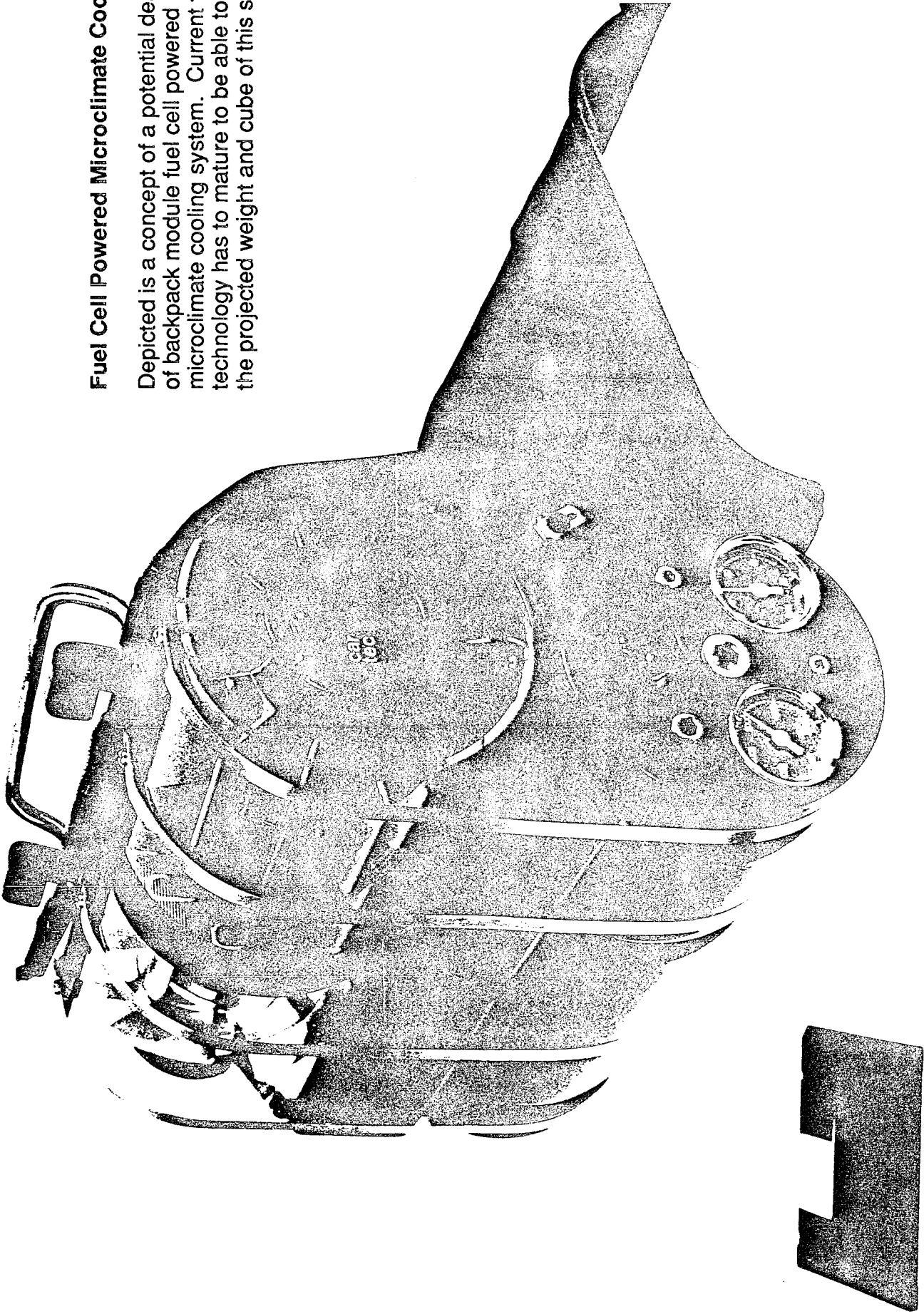
Project Goals
Size: 0.3 cu ft
Weight: 10 lbs
Duration: 4 hrs
Cooling: 300 W

Carlson Tech, Inc
Livonia, MI 48152
(313) 476-0013



Fuel Cell Powered Microclimate Cooling

Depicted is a concept of a potential design of backpack module fuel cell powered microclimate cooling system. Current fuel technology has to mature to be able to meet the projected weight and cube of this system.



Computer diskette (3 - 1/2 ")

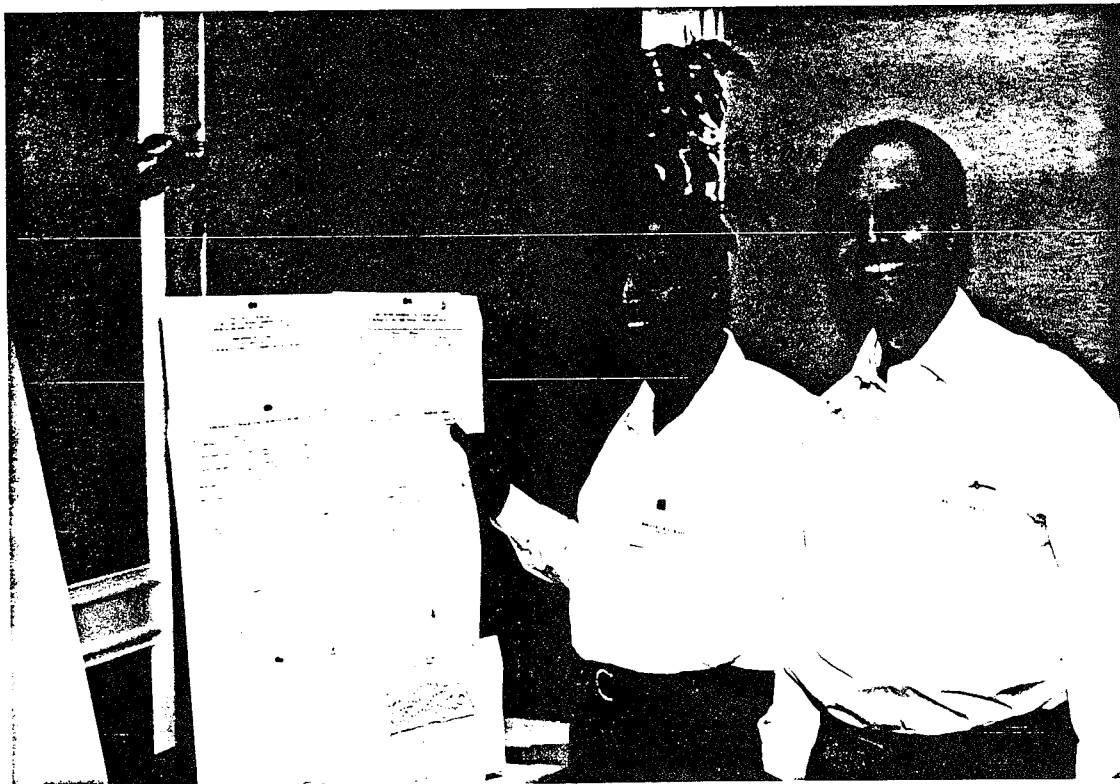
POSTER SESSION C

FACTORS GOVERNING
ON-LINE HYDROGEN GENERATION
FROM ALUMINUM IN ALKALI SOLUTION


BHASKARA M.L. RAO
EXPLORX INC.

6 FINCHLEY COURT, SOUTHAMPTON, NJ 08088

On-line generation of Hydrogen is of interest as fuel source for portable Hydrogen-oxygen Fuel Cell power source. Reactions associated with metal hydrides, hydrocarbon reformer and corrosion of light metals serve as the basis for developing the hydrogen generators. In this work, the factors governing on-line hydrogen generation using corrosion of aluminum in alkaline solution are elucidated using a laboratory reactor producing 0.15 Lpm of hydrogen. A major impedence to the production of hydrogen is that the aluminum surface is easily passivated. The surface passivation can be reduced by increased temperature and increased alkali concentration.



HYDROGEN STORAGE AND GENERATION METHODS

Method	Reaction/Condition	Basis
Compressed Gas	High Pressure 15-200 Atm	Storage
Liquid Hydrogen	Low Temperature Attrition: 1%/day	Storage
Reformer	$CH_3OH + H_2O = CO_2 + 3H_2$	On-line
Metal Hydride	$CaH_2 + 2H_2O = Ca(OH)_2 + 2H_2$ FeTiH ₂ + Heat FeTiH _x + x/2H ₂	On-line Reversible Storage
Aluminum Corrosion	$2Al + 6H_2O = 2Al(OH)_3 + 3H_2$	On-line 

RELATIVE RANKING OF HYDROGEN STORAGE & ON-LINE PRODUCTION METHODS

Method	Density (g/cc)	Weight (%)	Cost (\$/kg)	
			(A)	(B)
Aluminum Corrosion	1	2	1	2
Methanol Reformer	2	1	4	1
Calcium Hydride	2	3	4	4
FeTiH ₂	3	4	5	5
Cryogenic H ₂	4	1	2	1
Compressed gas	5	5	3	3

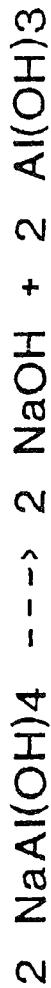
RANKING ORDER:

A: Raw Material + Installation Cost

B: Raw Material Cost

1 - Most Efficient
5 - Least Efficient

ALUMINUM-BASED H2 GENERATOR



Mass Fraction = 3.7

BACKGROUND

PREVIOUS WORK: (BATCH PROCESS)

Low Grade Al Scrap + NaOH Solution

-----> H₂ for Weather Balloons

THIS WORK: (CONTINUOUS PROCESS)

Al Scrap + NaOH Solution

-----> On-line generation of

H₂ for H₂-O₂ Fuel Cell.

Automobile Al Scrap:

450,000 tonnes/yr

= 50,000 tonnes/yr

= 2.5 MB Gasoline per annum

EXPERIMENTAL PARAMETERS

Type of Al Scrap	Composition Size, Shape
Reaction Control	Alkali Conc. Temperature Frothing Start/Stop
H2 Product Rate	ml/min/cm ²
Stoichiometry	Al Utilization Water Requirements
Reactants/Products Management	Al & Water Addition Solids Conc./ Removal H2 Purification Safety

MATERIALS

ALUMINUM

2.5 mm dia. * 6 mm Wire Scrap

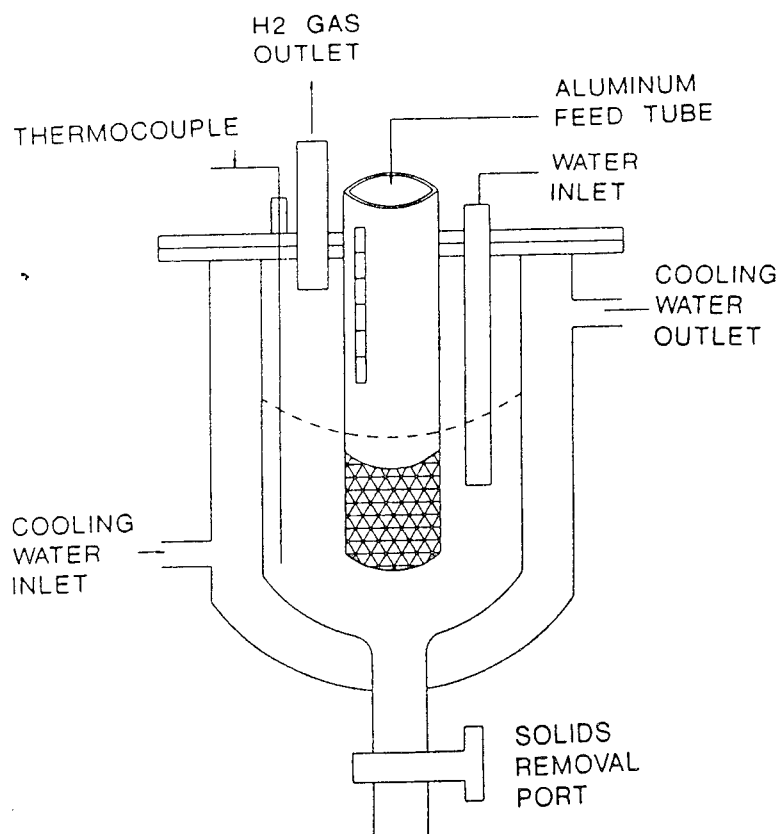
7 cm²/gram

99.7% Aluminum

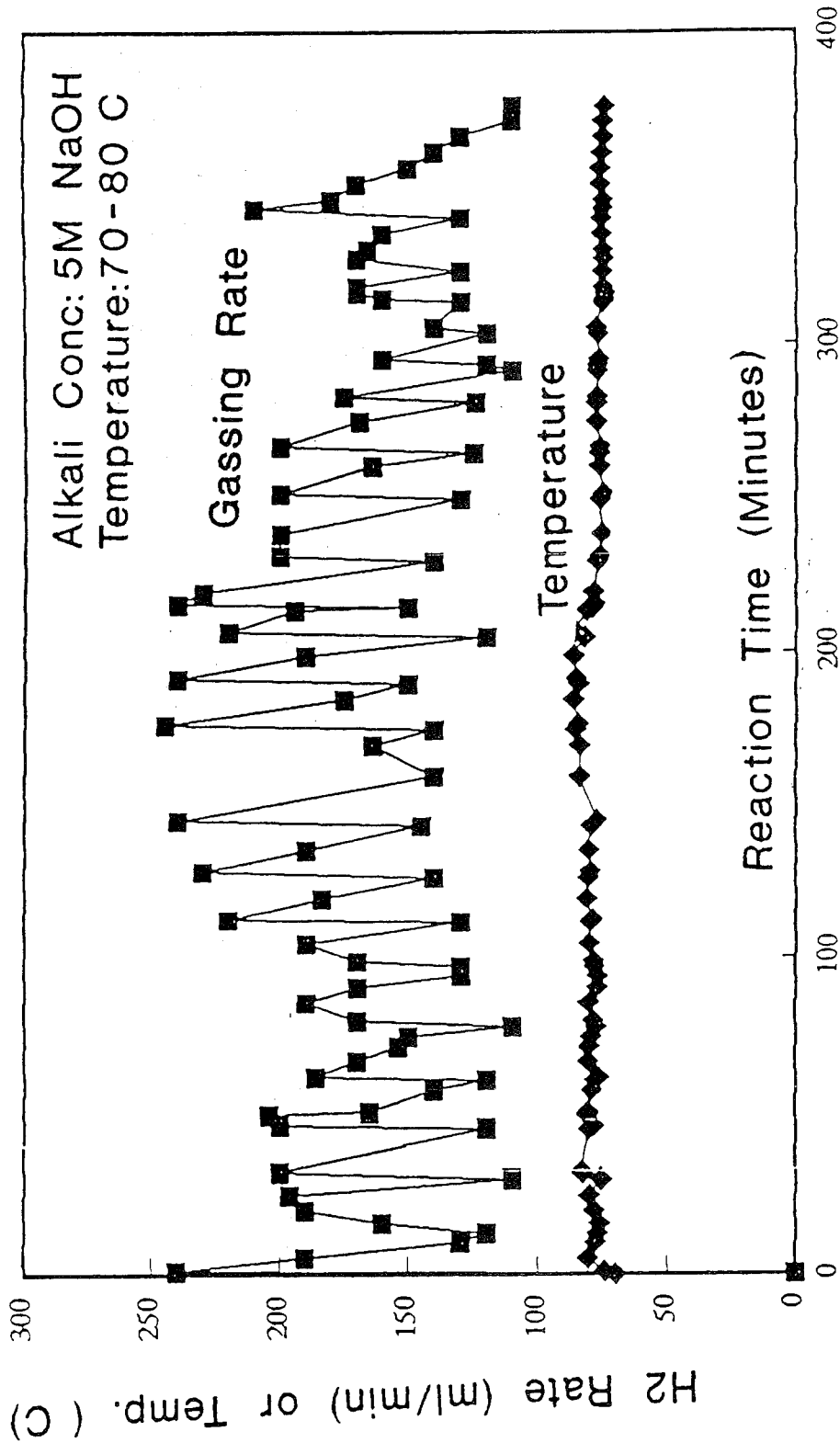
ELECTROLYTE

NaOH (2-5M) With Al(OH)₃ Pptn.

EXPERIMENTAL SET-UP



150 ml/min H₂ Generation
With CAWS + Water Additions



Rate Decreases as Al(OH)₃ ppt. ~ 50 wt%
1.5x Water Needed To Maintain Rate

SUMMARY OF DATA COLLECTED FROM 5 Lpm HYDROGEN GENERATOR

1. PREFERRED ALUMINUM SOURCE - CHOPPED AL WIRE
2. CORROSION SOLUTION - 5.75 M NaOH
3. TEMPERATURE - 85-90 C
4. STARTING VOLUME - 333 mL NaOH PER Lpm H₂
5. H₂ PRODUCTION RATE - AVERAGE 4.6 mL/min.cm²
6. START-UP - SELF HEATING 20-90 C
7. RESTART - REQUIRED STIRRING AND WATER ADDITION
8. SHUT DOWN - RESIDUAL RATE, $1 * 10^{-3}$ TIMES THE FULL PRODUCTION RATE
9. PRECIPITATION - YES
10. FOAMING - YES

POSTER SESSION D

SHORT ABSTRACT

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

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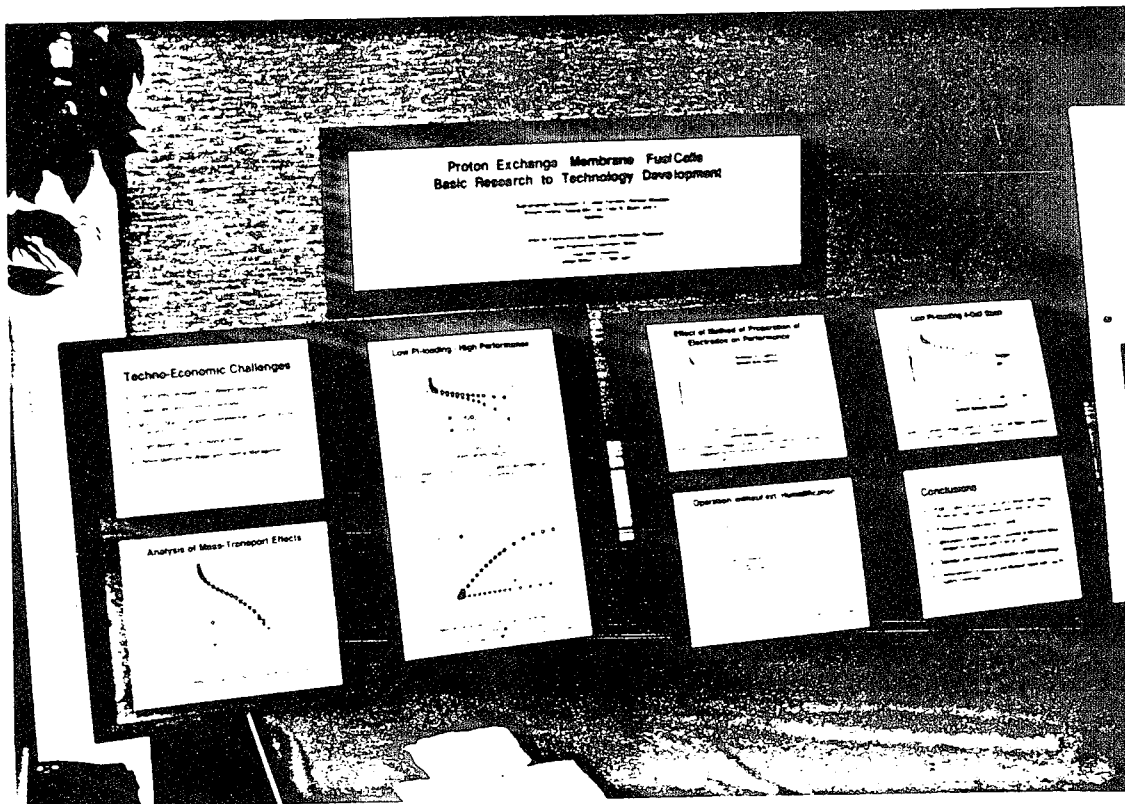
Since the late 1950s, there have been three incentives for the development of fuel cell power sources - the NASA Space Program, initiated in the late 1950s; the Energy Crisis in 1973; and the Environmental Problems and legislations in the 1980s and 1990s. In view of the abundance of natural gas resources and the good prospects for its direct utilization in the high temperature fuel cells, the focus of the fuel cell program is based on natural gas or natural gas derived fuels. The proton exchange membrane fuel cell (PEMFC) is one of the most promising candidates for space, electric vehicle and portable power sources. The rationale for this statement is that, from an electrode kinetic point of view, it shows the best prospects for attainment of the highest energy efficiencies and power densities. The presentation material highlights the following accomplishments at Texas A&M University, which covers the spectrum of basic research to technology development:

- (i) development of a micro electrode technique to determine electrode kinetic, mass transport and ohmic parameters;
- (ii) optimization of the structures and compositions of the electrodes and of membrane and electrode assemblies to lower platinum requirements (to 0.2 g/kW) and attain higher energy efficiencies and power densities;
- (iii) demonstration of low ohmic losses with advanced membranes having high water content and good water retention characteristics;
- (iv) experimental and theoretical analysis which provided an insight on methods to minimize mass transport limitations with air as a cathodic reactant and finding an empirical equation, which excellently fits the experimental cell potential vs. current density data;
- (v) finding solutions to the challenging water and thermal management problems - for example (a) operation of PEMFCs without external humidification, and (b) evaporative/air cooling;
- (vi) multicell stack development with low platinum loading electrodes; and
- (vii) experimental analysis of PEMFCs illustrating the electrocatalytic and depolarization problems in PEMFCs using methanol directly as a fuel.

The techno-economic challenges in PEMFC technology are as stressed in the relevant viewgraph material.

PROTON EXCHANGE MEMBRANE FUEL CELLS - BASIC RESEARCH TO TECHNOLOGY DEVELOPMENT

Poster Session



WORKING GROUP SUMMARIES

In this section, the reader will find a summary prepared by the individual Working Groups, followed by a complete set of the presentation materials briefed in the final Plenary Session. In that final session, each Chairperson briefed the Workshop participants on the results of the Working Group discussions and presented the Findings and/or Recommendations of that particular group.

WORKING GROUP I SUMMARY

PROSPECTOR VII: SMALL, PORTABLE FUEL CELLS**GROUP A****APPLICATIONS FOR COMMERCIAL PRODUCTS & SPECIFIC MISSIONS****INTRODUCTION**

The Applications for Commercial Products & Specific Missions Working Group (A) was tasked with the identification and analysis of applications for small fuel cells with output power less than 1kW. The charge to the working group included the following requirements and constraints:

- Small fuel cells means less than 1 kW at the upper end of the range and as small as is practical at the lower end.
- The military will always be looking for better power sources, but the current emphasis is on the Soldier System.
- The complete system has to be carried; it is the energy density of complete systems that counts.
- Assess the state-of-the-art of low power (< 1 kW) fuel cell systems.
- Assess whether it is reasonable to try to develop much smaller fuel cell systems for battery replacement, i.e. assess the scaling laws, heat transfer, fuel supply, etc. issues.
- Address some system issues such as: (i) stack integration, (ii) air quality/clean-up, (iii) environmental effects (e.g. freeze/thaw, etc.), (iv) attitude sensitivity, (v) shelf-life, readiness, (vi) start-up time, simplicity of shut-down.

WORKING GROUP PARTICIPANTS & RATIONALE

The working group participants, listed in table 1, were drawn from military, industrial and academic organizations, allowing a broad perspective of fuel cell applications. Essentially, "users," "technologists," and "appliers," were represented from government, university and industry organizations. There was a heavy prevalence of industry personnel (50%) allowing the development of applications to be studied from a commercial-manufacturability standpoint. Government and university representation was 33% and 17%, respectively. During later discussions the group was augmented by the presence of extra government personnel, mainly from the Army.

To establish the ground rules for the discussion, the chairman provided a review of the workshop requirements in terms of the information to be assessed and the areas in which conclusions should be reached by this working group. The group initiated its discussion by a statement from each participant outlining their goals and expectations for this workshop. While these reflected the organizational backgrounds of the individuals concerned, a significant correlation of positions, none conflicting, evolved. The group then considered the planned discussion topics, determining the relevant questions to be addressed by the group, and deciding upon the format of

G	Tom MALONEY	NYMA Inc. / NASA Lewis Res. Center	[chair]
A	David HILL	Auburn University	[assist. chair]
G	Narayan DODDAPANENI	Sandia National Laboratories	
I	Sandors ABENS	Energy Research Corporation	
G	Kathy HITHE	Department of Defense	
I	Radha JALAN	Electrochem	
I	Richard VARJIAN	Dow Chemical	
I	Jerry CHODIL	Ball Aerospace	
I	James HUFF	Ballard Power	
A	Frank ROSE	Auburn University	
I	Mohammad ENAYETULLAH	H-Power Corporation	
G	Walter TASCHEK	US Army Belvoir RD&E Center	

Table 1. Participants in the Applications for Commercial Products & Specific Missions Working Group. A = academic; G = government; I = industry.

the final report and briefing to the Workshop body. It was decided to use the three formal questions put to the working group by the workshop secretariat as the framework for discussion. Also, no discrimination was made between applications and Army missions.

Below are included a series of introductory quotes from the various members of the working group, representing their fundamental view of the workshop and what they hoped to obtain from their participation. It should be noted that there was a broad range of users, technologists, appliers, etc. from industry, the military, government, and universities.

- Narayan Doddapaneni "...interested in applications of small motors of all types."
- Sandors Abens "...representing a company whose principal business is in high energy fuel cells since 1969. Has produced many US Army tactical powerplants. Major thrust is in direct hydrocarbon fuel cells with a 2 MW demonstration occurring in 1995. In the 3-15 kW range, is developing push-button, microprocessor-controlled fuel cell systems using methanol with external reforming and having conditioned power output with load-following capability. This represents relevant experience for the Soldier System."
- Kathy Hithe "...fuel cell program manager developing small, man-portable power systems for special operations forces. Performance and reliability are the top priority over leave-behind or throwaway systems..."
- Radha Jalan "...small business owner involved in product development to increase the volume of sales of fuel cells. Wants to see fuel cell technology become integrated with household products. Market potential depends on public information. "
- Richard Varjian "...representing a supplier of membranes and electrode systems to major fuel cell manufacturers. Very interested in PEM fuel cell applications for both military and commercial activities. The scale-down problem is of crucial interest in such applications and also the use of either hydrogen or methanol fuels."
- Mohammad Enayetullah "...running a fuel cell hybrid bus program and using small PEM-based fuel cells in the 10-100mW range with air-breathing mode at 30-50 ASF."
- Walter Taschek "...working on the integration of the Soldier System micro-climate cooling (MCC) system with fuel cell-based power systems. Deep background in low power, long mission systems."

- James Huff "...working on PEM-based fuel cells since 1983 for utility and transportation (e.g. buses) organizations. Has tested a 6-cell, 25W system for 25,000 hours to demonstrate small, long-life systems."
- Frank Rose "...interested in the key issues of fuel cell science and technology to solve problems associated with a broad range of military and commercial power requirements. Also interested in competitor technologies. Sees the need for universities to work with industry and to licence patents to industry, making money for both the industrial and academic partners."
- Jerry Chodil "...representing a power systems integrator to take what is available and to incrementally improve the elements to provide a better total system. Especially interested in the issues of reactant storage. Wants to find the right niches for products in the government procurement market."

GENERIC ARMY REQUIREMENTS

Three major Army infantry missions where a fuel cell may be employed on the soldier to provide autonomous power were considered; (i) Dismounted Infantry Combat, (ii) Airborne Deployment, (iii) Special Operations. All three have different logistics requirements for fuel supply. Such missions are represented, in terms of peak power and energy storage requirements, by the typical missions listed in Table 2. Note that energy storage value is based upon mission's duty cycle.

Duration	Power - Watts (W)	Energy - W-Hrs
4 Hours	300	1,200
6 Hours	30	180
8 Hours	100	800
24 Hours	100	2,400
24 Hours	500	2,000
48 Hours	250	12,000
72 Hours	300	5,000
7 Days	100	700
30 Days	50	3,000

Table 2. Projected power and energy requirements for a broad-range of typical Army missions. Note that the duty-cycle is different for the various missions. [source: Walter Taschek, US Army Belvoir RD&E Center, VA]

In terms of mission repetition rate, any FC-based power system will have to be able to operate over dozens of missions (i.e. ~24 to 60) before requiring major overhaul, resulting in a lifetime of between 500 and 1,000 hours, typically. Also, such a system may have to operate over 100 hrs (i.e. several

short missions) under extreme environmental conditions (e.g. dust, temperature, rain, etc.) without access to maintenance facilities other than those that can be provided in-the-field by combat soldiers. This set of requirements far exceeds the demands of ordinary commercial systems.

STATE-OF-THE-ART ASSESSMENT

Although it was not explicitly defined by the working group, the initial discussions did generate a generic system configuration that provided the basis for all discussions. Such a generic system comprises the following elements:

- Converter (stack)
- Fuel tank
- Oxidizer tank (optional)
- Fuel (and oxidizer) delivery sub-system
- Thermal control sub-system (incl. fan, filter, etc.)
- Electrical output power conditioning
- Structure (other than that integral to the other elements)

System-level requirements added by the working group included:

- Fuel cell elements should be re-usable
- Fuel pack should be replaceable/exchangeable for fast refueling
- System operation should be attitude insensitive
- Fuel and oxidizer supply should have high purity to support reliability and lifetime

When discussing the applicability of fuel cells to Army missions and other commercial products there were several metrics which figured in the discussions:

- Peak power
- Energy available (i.e. fuel capacity, fuel consumption, conversion efficiency, etc.)
- Weight
- Volume
- Reliability
- Stack (cell) output voltage
- Output current

Subject to the Army requirements, the H₂-Air PEM fuel cell system using forced air cooling was baselined as state-of-the-art. Compressed hydrogen storage was assumed for the baseline because of the inherent simplicity of supply, the immediate multiple start/stop, freeze/thaw capability, and

attitude insensitivity requirements. No oxidizer tank is necessary for this configuration. The following specification for a Soldier System H₂-Air PEM fuel cell stack operating at 1 atm was determined to be state-of-the-art:

- 150 W output electrical power
- H₂-ambient air w/filtration
- Forced ambient air convective cooling
- Efficiency ~50% @ 140 ASF to 65% @ 80 ASF
- Weight ~5 - 7 lbs (3.6 kg - 4.5 kg)
- Volume ~150 - 170 cu.in. (~2.5 ltr)
- Lifetime > 500 hours with proper care (i.e clean fuel and filtered air supply)

Stack mass was estimated to be approximately 20 - 30 W.lb⁻¹ (= 44 - 66 W.kg⁻¹) peak power using Nafion® 117 membranes. The working group agreed that moving to Nafion® 105 membranes would improve the power conversion efficiency by upto 50% taking the specific power to the 30 - 45 W.lb⁻¹ range. Stack mass for the baselined 150W electrical power requirement results in a 5 - 7 lb stack using Nafion® 117 with a reduction to the range 3.3 - 5 lb for Nafion® 105.

Then, state-of-the-art PEM FC-based systems were analyzed in terms of mission duration, energy storage, all-up system weight and volume, and hydrogen usage. These data are shown in table 3

Energy [W.hr]	Duration [hr]	H ₂ Mass [kg]	Tank Mass/Vol [kg] / [ltr]	Total Mass/Vol [kg] / [ltr]	W.hr.kg ⁻¹	W.hr.ltr ⁻¹
900	6	0.050	0.93 / 0.92	1.00 / 0.92	130	240
1200	8	0.064	1.17 / 1.20	1.20 / 1.20	165	300
1800	12	0.100	1.70 / 1.70	1.80 / 1.70	230	400

Table 3. H₂-air FC-based power system operating at 150 W output power. FC stack has the following characteristics: current density = 100 ASF, efficiency = 60%, weight = 4.5 kg, volume = 2.5 ltr. H₂ tank P_bV/W = 1.1E6 in., safety factor = 4. Ancilliary systems are estimated to weigh 1.5 kg with a volume of 0.3 ltr. Based on a H₂ burn rate of 0.0565 kg.kW⁻¹.hr⁻¹ (= 0.1243 lb per kW-hr)

Reversible and primary hydrides may be feasible as fuel supplies, pending further development, especially for commercial applications. Other technologies such as methanol-air/O₂

fuel cells and solid oxide fuel cells were not thoroughly assessed since those technologies are not yet mature and cannot be assessed at a system level.

Balance-of-plant elements (fuel delivery system, thermal control system, power conditioning, structure) are not regarded as significant problems for FC-based power systems. The technologies required for these sub-systems are well-developed. Mass and volume clearly must be minimized, but this can be accomplished by iterative design improvements and weight reduction programs. For example, structural mass can be reduced by substituting Al-Li alloys or composite materials for typical aluminum structural members.

Analytic Power Corporation presented a state-of-the-art small fuel cell system (FC-150) with the following system specifications:

Power	153 Watts	Voltage	28 Volts
Current	5 Amp	Cell Voltage	0.723 Volts
Current Density	125 ASF	Number of Cells	39
Length	11.5 in.	Width	4 in.
Height	5.75 in.		
Weight	8.1 lb.	Volume	265 cu.in.

H₂ gas storage under 10,000 PSI pressure was considered state-of-the-art using a Kevlar-wrapped tank. Data provided in a report by Directed Technologies, Inc. ["High Pressure Hydrogen Supply for Increased Fuel Cell Efficiency," James, B.D. et al., Phase I Final Technical Report, US Army Research Office contract DAAH04-93-C-0005, Oct. 1993] indicated that some commercially-available storage tanks are available, some meeting MIL-STD-1522A requirements.

Directed Technologies' analysis shows that Structural Composites Industries (SCI) produces two composite fiber wrapped tanks (AC-5184 and AC-5128A) capable of being used for Soldier Systems applications. Both tanks are ~22" long with a diameter ~6.5" and weigh 8.8lbs and 6.6lbs respectively. Assuming an energy based on H₂/air fuel cell burn rate of 0.124lbs/kW-hr (200ASF, 0.7V/cell fuel cell stack) the AC-5184 tank could provide ~5.2kW-hr energy storage whereas the AC-5128A tank could provide ~4.5 kW-hr, both at a safety factor of 1.5 under a pressure of ~8,000 PSI.

Directed Technologies also presented two advanced designs for 15,000 and 10,000 PSI tanks providing 5 kW-hr energy storage in tanks of length 16.7" with 5.4" and 6.3" diameters, respectively. It must be noted, however, that a current density of 200ASF (amp/sq ft) is a near-term development goal and that ~125ASF is the state-of-the-art.

The state-of-the-art for fuel delivery and management systems was not clear to the working group except to say that weight is estimated to be ~1 lb. As a result, the working group was of the opinion that the 1996 near-term goals set for an integrated system with MCC requiring 900 W-hr energy storage in a 7 kg system can be met from the current baseline. Significant effort will have to be expended to reduce system mass contributions (e.g. structures, etc.) and also to minimize storage tank mass. With regard to the 1998 requirements of 1.2 kW-hr energy storage within a 5 kg system the achievability is less well-defined but is certainly not regarded as impossible using H₂-air PEM fuel cells for power generation.

ASSESSMENT OF REASONABLENESS TO DEVELOP SMALLER FUEL CELL SYSTEMS FOR BATTERY REPLACEMENT

One major consideration for the working group was whether very small fuel cells can be developed to replace batteries. For comparison, the BA-5590 (Li/SO₂) and BA-6590 (Li/SOCl₂) single D-cell battery data* have been included here in the 20-24W range (i.e. current discharge of 8A):

Type	Voltage [V]	Capacity [A-hr]	*Weight [lb]	Specific Power	Energy Density
BA-5590	2.5	6.7	0.18	111 W.lb ⁻¹	94 W-hr.lb ⁻¹
BA-6590	3.0	11.7	0.26	92 W.lb ⁻¹	134 W-hr.lb ⁻¹

Likewise, below is presented data for multiple D-cell based battery packs using the same technology and using a discharge rate of 2A:

Type	Voltage [V]	Capacity [A-hr]	Weight [lb]	Specific Power	Energy Density
BA-5590 10 cells/pack	28	7.5	2.2	25 W.lb ⁻¹	94 W-hr.lb ⁻¹
BA-6590 8 cells/pack	28	12.5	2.25	25 W.lb ⁻¹	156 W-hr.lb ⁻¹

Note that the extra weight for the multiple cell power packs is attributed to packaging mass. Data were obtained from Prospector III - High Energy Density-High Power Density Power Sources R&D Workshop Report, pp341-342, May 1992.

The working group agreed that there was nothing intrinsically wrong with considering scale-down of fuel cells to the typical battery range as indicated by the data above. However, it was the opinion of the group that there is a major concern over scale-down due to voltage and current

requirements. For example, a 20W FC-based power source could comprise a single cell operating at 0.7V. Alternatively, such a 20W source operating at 24V would result in 34 very small cells, possibly of an impractical size for manufacturing. Conversely, stepping up 0.7V output from a single cell operating at 20W to 24-28V output would require DC-DC power conditioning elements.

Further questions arose over whether very small fuels cells are competitive on a mass and volume basis. It was determined that not all fuel cell system elements scale similarly. For example, fuel storage and delivery systems become more massive relatively-speaking as the system size is reduced. Larger systems benefit from structural economies-of-scale. Without adequate data from actual small fuel cell systems operating at very low power levels no specific recommendations could be made except to say that research in this area is required to determine if such concerns are completely valid. However, the working group was of the opinion that a fuel cell power source can be used as a battery-charger while not powering the Soldier System MCC and so batteries should be retained, in the Soldier System, to power the non-MCC sub-systems. Power and energy efficiency may be reduced by such an approach, but operability would be significantly enhanced.

SYSTEM ISSUES AND RISKS

The working group was of the unanimous opinion that cell structure and manufacturability are very dependable and that operation over periods in excess of 10,000 hours is proven, dependent on the purity of the reactants. In this case, air filtration was considered absolutely necessary. If logistic fuels such as JP-8 (diesel) are to be used via a reformer, the degree of sulfur contamination is extremely critical for long-life and reliable operation. The working group also considered that FC-based systems will have long shelf-lives if suitably packaged (i.e. water vapor contamination is minimized).

With regard to freeze-thaw tolerance, the working group believes this to be adequate with some fuel cell unit tests indicating no performance degradation after eleven (11) cycles over 2,500 hours. Essentially, the problem caused by freeze-thaw cycling is that of expansion-contraction of water (ice) in the membrane structure. Dow Chemical has indicated that PEMs will survive in excess of 100 freeze-thaw cycles with no degradation in membrane characteristics. One further problem area identified as a result of these discussions was that PEM fuel cells will not start at temperatures below freezing. Therefore, some heating capability will be necessary for sub-zero start-up.

There was some discussion on the issue of shock sensitivity and the impact of projectiles onto FC components and peripheral sub-systems. Some concern was expressed at the safety of high pressure H₂ gas tank penetration due to the explosive nature of H₂ in air. It was pointed out that shrapnel from modern fiber composite wrapped tanks is less than that generated by all-metal vessels. A major question to be answered is how much kinetic energy is required to initiate an explosion if a compressed GH₂ tank is ruptured under ambient atmospheric conditions. Further discussion raised the issue of soldier wounds due to tank shrapnel. This is regarded as much less life-threatening when compared to a wound produced by a bullet that has penetrated a Lithium battery and carried Lithium compounds into the soldier's body tissue. The group determined that impact testing should be initiated if it has not already been done.

The problem of using compressed GH₂ leads to the issue of user acceptance for fuel cell systems which the working group believed must be determined soon. For some applications, such as airborne soldier deployment, the proper certifications must be obtained. Clearly, acceptance can only be achieved if safety under operational conditions can be demonstrated. The results of technical analysis cannot be substituted as a means to obtain user acceptance and in this case the end user is an individual infantryman in combat.

At a higher level, user acceptance will be dominated by the logistics of deployment. The working group believed that the entire fuel supply scenario should be determined. Many questions arose: Where is the fuel produced? How does the supply chain work? Are there any disposal requirements? Cost? Safety? The group raised the issue of what types of field generators should be used, e.g. diesel reformers, diesel-generator powered water electrolyzers. A further question was raised: Can a squad-level H₂ generator be mounted on an M2 Bradley Fighting Vehicle? For dismounted missions a nine-man squad requires $9 \times 0.62\text{lbs} = 5.6$ lbs of hydrogen per 5 kW-hr autonomous mission and only ~ 2.7 lbs of hydrogen per 2.4 kW-hr mission. However, an H₂ compressor/bottling system would also be required to complete the logistical system. The hydrogen fuel supply logistics needs serious consideration and there are several alternatives, although it will probably not be as difficult as the Lithium battery re-supply logistics.

The fuel cell system should be fully integrated with the micro-climate cooler and it must be designed for easy maintenance (i.e., easy air filter replacement). The forced air cooling fan should present no serious problems since the audio and electrical signatures are insignificant compared to the soldier system refrigeration compressor. If needed, the fuel cell stack can be operated at low power density to relieve the cooling duty. In fact, very small power systems may need to be heated, rather than cooled. Accurate designs of very low power (i.e., 20W) fuel cell systems can be achieved, but feasibility will be highly dependent upon the system current and voltage requirements. In terms

of system risks, the air breathing fuel cell may be subject to contaminated environments from dusty air and chemical warfare. It may be necessary to include a small oxygen/air supply for emergency circumstances. Long term biological growth must be prevented.

COMMERCIAL APPLICATIONS

Military applications can benefit from economies of scale generated when commercial applications become feasible. To get such benefits a "virtuous circle" needs to be set up. Technology transfer from military R&D projects along with its associated "political benefits" represent the flow from military to commercial sectors while economies-of-scale, lower procurement costs, and follow-on commercial R&D represent the flow from commercial to military sectors.

For fuel cell based power systems cost will be the most important issue for commercial acceptance. Niche markets will be established for the commercial low power systems. Investment toward an independent market survey may help to identify some of these markets and possibly lead to dual use cost share. On the other hand, a customer base will emerge if a dependable, affordable product is available. One critical area will be the supply of hydrogen to consumers under safe and reliable conditions subject to OSHA and NFDA regulations. If fuel cell based power sources which are directly applicable to the Soldier System scenario can be applied in the commercial sector then massive savings can be made in procurement.

Some potential commercial markets that were identified included:

- Camera power pack for TV crews
- Remote radio receivers
- Remote lighting and emergency lighting
- Wheelchairs, golf carts, etc.
- Lawn tools, hand tools, construction tools
- Luxury RVs where cost is tolerable
- 500W-level precise power (i.e. computer power)
- Uninterruptible power supplies (especially where stringent noise levels prevail)
- Cellular power and repeater stations

Finally, the working group was of the opinion that the commercialization dynamic may best be summed up in the phrase "Build it and they will come." In this case, military R&D and applications will lead the way for commercial applications.

Thomas MALONEY **NASA LeRC** **[chair]**
David HILL **Auburn** **[asst. chair]**

Narayan DODDAPANENI **Sandia**
Sandors ABENS **Energy Res Corp**
Kathy HITHE **DoD**
Radha JALAN **Electrochem**
Richard VARJIAN **Dow Chemical**
Jerry CHODIL **Ball Aerospace**
James HUFF **Ballard**
Frank ROSE **Auburn**
Mohammad ENAYETULLAH **H-Power**
Walt TASCHEK **Belvoir**

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OVERVIEW

- Began with discussion of low power fuel cell state-of-the-art
- Defined reference system (H₂/air PEM fuel cell)
- Studied reference system

Group A

State-of-the-Art PEM H₂/air Fuel Cell Stack - 1

- α 150We
- α H₂-ambient air
- α Forced convection cooling
- α Efficiency ~ 50 - 65%
- α Current density ~ 140 - 80 ASF [A.ft⁻²]
- α Mass ~ 8 - 10 lbs
- α Volume ~ 150 - 170 cu.in (~ 2.5 ltr)

Reliable with sufficient life (7500 hrs) "*If treated right*"

- α multiple start-stop capability (instantaneous)
- α freeze-thaw tolerance is adequate
- α long shelf-life in ready state

Group A

State-of-the-Art PEM H₂/air Fuel Cell Stack - 2

- α Rugged if shock mounted
- α Attitude sensitivity dependent on balance-of-plant (e.g. fuel delivery) and possibly in-stack fluid

Balance-of-Plant

- α Fuel Delivery System
- α Thermal Control System
- α Electrical Output Power Conditioning
- α Structure
- α Mass and volume are system (application) specific

Group A

- H2 Gas Storage
 - α 10,000 PSI pressure with Kevlar-wrapped tank
 - α Safety factor = 2.2
 - α Figure of merit = $P_B \cdot V/W = 1.1e6$ in
 - α 800-1000 W-hr/kg

- Reversible Hydrides
 - α Thermal losses present system-level problems in thermal control and management
 - α Start-stop issues (slower start; difficult to stop/re-start)
 - α 400 W-hr/kg (400 W-hr/ltr)

- Primary Hydrides
 - α System-level implications; water; thermal
 - α Start-stop issues
 - α 1700 W-hr/kg

State-of-the-Art PEM Fuel Cell Systems - Mission Time Breakdown

150 We	6 hrs	900 W-hr	7 kg	3.3 ltr	130 W-hr/kg	290 W-hr/ltr
150 We	8 hrs	1200 W-hr	7.3 kg	3.5 ltr	165 W-hr/kg	340 W-hr/ltr
150 We	12 hrs	1800 W-hr	8 kg	4 ltr	225 W-hr/kg	450 W-hr/ltr

ARMY TARGETS

150 We	FY96	130 W-hr/kg	(6 hr operation)
150 We	FY98	240 W-hr/kg	(8 hr operation)

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Group A

- PEM Fuel Cells Should Be Considered for Battery Replacement
 - α There are many operational and logistics issues to consider
 - α Replace some or all of the soldier's batteries?
(technically feasible - but are FCs competitive on a mass and volume basis?)
 - α Use FCs as battery charger when not powering MCC

- Scaling Laws
 - α Accurate designs can be performed
 - α Stack design is highly dependent on current and voltage requirements

Heat Transfer

- α Cooling fan noise and electrical signature are insignificant compared to soldier system refrigeration compressor
- α If possible, operate at low power density to relieve cooling duty
- α Very small power systems may need to be heated, not cooled

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Group A

H2 Fuel Supply Issues

- α **Must consider entire fuel supply scenario**
 - where it comes from
 - how it is disposed
 - safety
 - cost
 - supply chain

- α **Logistics need serious consideration, but probably not as difficult as Li battery re-supply**

- α **Use of field generators: what types?**
 - What level? (battalion, company, platoon, squad)
 - Can squad-level generator be mounted on M2 BFV?

Commercial Low Power H2-air PEM Fuel Cell Applications

- α Cost dependent
- α TV camera power pack for TV crews
- α Remote radio receiver
- α Remote lighting & emergency lighting
- α Wheelchairs, golf carts
- α Lawn tools, hand tools
- α Luxury RVs where cost is tolerable
- α 500W level, 400Hz precise power (i.e. Patriot missile)
- α Uninterruptible power supplies

*Commercial re-supply logistics is burdensome or not available,
maybe re-chargeable hydrides are acceptable*

*Market survey may identify markets and lead to dual use cost share
"Build it and they will come"*

WORKING GROUP II SUMMARY

INTRODUCTION

The Alternative Fuel Cell-Related Technologies Working Group (Group B) was tasked with identification and analysis of alternative fuel cell technologies, as well as assessing alternatives to fuel cells as a technology, and identifying and characterizing fuel cell options with respect to size. The notion of a disposable fuel cell system was to be addressed, and the state-of-the-art was to be summarized, if possible, for small fuel cells with output power of less than 1kW. The sponsor's detailed Charge to the Working Group included the following requirements and/or constraints:

- Identify *alternatives to fuel cells* in small size ranges, and provide a brief summary of the status of each to include its range of applications.
- Assess *alternative fuel cell technologies* for systems under 1kW, and summarize the advantages and disadvantages of each.
- Identify and characterize (prioritize) low power fuel cell systems, specifically "self-contained" options with respect to their size and/or range of application.
- Assess whether it is reasonable to develop the notion of disposable fuel cell systems for certain missions.
- Summarize the state-of-the-art of low power (< 1 kW) fuel cell systems.

Guidelines and definitions for Group B discussions included these points:

- "Small fuel cells" or "low power fuel cells" are defined by the sponsor to be those systems which are less than 1 kW at the upper end of the range and as small as is practical at the lower end.
- The Group should keep in mind that the military is looking for better power sources, but the current emphasis is on the Soldier System.
- The complete system has to be carried by the individual; it is the energy density of complete systems that counts.
- Make all comparisons on a "level playing field" basis.

WORKING GROUP PARTICIPANTS

The working group participants, listed in table 1, were drawn from military, industrial and academic organizations, allowing a broad perspective of fuel cell applications. Essentially, "users," "technologists," and "appliers," were represented from government, university and industry organizations. There was a heavy prevalence of government personnel (65%), allowing the development of applications to be studied from a "government applications" perspective, with emphasis on the Soldier System. Industry and university representation was 14% and 21%, respectively. It was anticipated that the group might finish its discussion early, due to the "established nature of the technology," and would then break into two parts to participate in the discussions of the remaining two groups during the late Wednesday evening session. This did not prove to be the case, as individuals found ample opportunity to weigh all fuel cell and non-fuel cell options in great detail against the criteria and constraints.

G	Robert HAMLEN	Army Research Laboratory	[chair]
A	Eugene SMOTKIN	Illinois Institute of Technology	[assist. chair]
G	Robert ROSENFELD	ARPA	[assist. chair]
I	Bhaskara RAO	Explorex	
G	Rajiv DOSHI	Argonne Laboratories	
G	Jay STEDMAN	w/ARPA	
G	Mark GRAF	Department of Defense	
A	Donald LIN	Indiana - Purdue	
G	Pete HOWARD	US Army Belvoir RD&E Center	
G	Brad LAPRISE	US Army Natick RD&E Center	
A	Ron MANN	Royal Military College of Canada	
I	Eric SIMPKINS	Energy Research Corporation	
G	Richard WELLER	Naval Surface Warfare Center	
G	Richard PAUR	Army Research Office - Sponsor	

Table 1. Participants in the Alternative Fuel Cell-Related Technologies Working Group (Group B). A = academic; G = government; or I = industry affiliation.

To establish the ground rules for the discussion, the chairman provided a review of the Workshop requirements in terms of the information to be assessed and the areas in which conclusions should be reached by this Working Group. The Group initiated its discussion by a statement from each participant outlining their goals and expectations for this Workshop. While these reflected the organizational backgrounds of the individuals concerned, a significant correlation of positions, none conflicting, evolved. The Group then considered the planned discussion topics which had been presented in the Charge by the sponsor, Dr. Richard Paur (ARO), determining the relevant questions to be addressed by the group. The format of the final report and brief-out to the Workshop body was determined. The Group decided to study and compare the many attributes of fuel cell systems, as well as their competitors, batteries and thermophotovoltaics, and report on their relative merit.

In the early portion of the discussion, there were seven fuel cell options and four non-fuel cell options identified to be examined by the Group. The fuel cell options were as follows: chemical hydride PEM, pressurized hydrogen gas PEM, physical hydride PEM, direct methanol PEM, and reformer with PEM. Additionally, in the logistics fuel regime, there were two more candidates, namely, the solid oxide fuel cell and the phosphoric acid fuel cell. The non-fuel cell options included the following competitors: aluminum-air batteries, zinc-air batteries, thermophotovoltaics (TPV) and alkaline-metal thermoelectric converter (AMTEC).

DISCUSSION

I. Portable and Mobile Power Systems

The Group discussed the various options among fuel cell systems and decided to categorize them into three main categories according to the source of the fuel/power for the power system itself, the portion of the sub-set of the system which would (or could) be located in a heavy logistics infrastructure ("heavy base"), and that portion of the power system which would be deployed forward to a remote site for various military missions or civil applications. Figure 1 illustrates a general map of those probable power systems and how they might be arrayed across the supply-heavy base-remote site logistics grid.

Portable and Mobile Power Systems

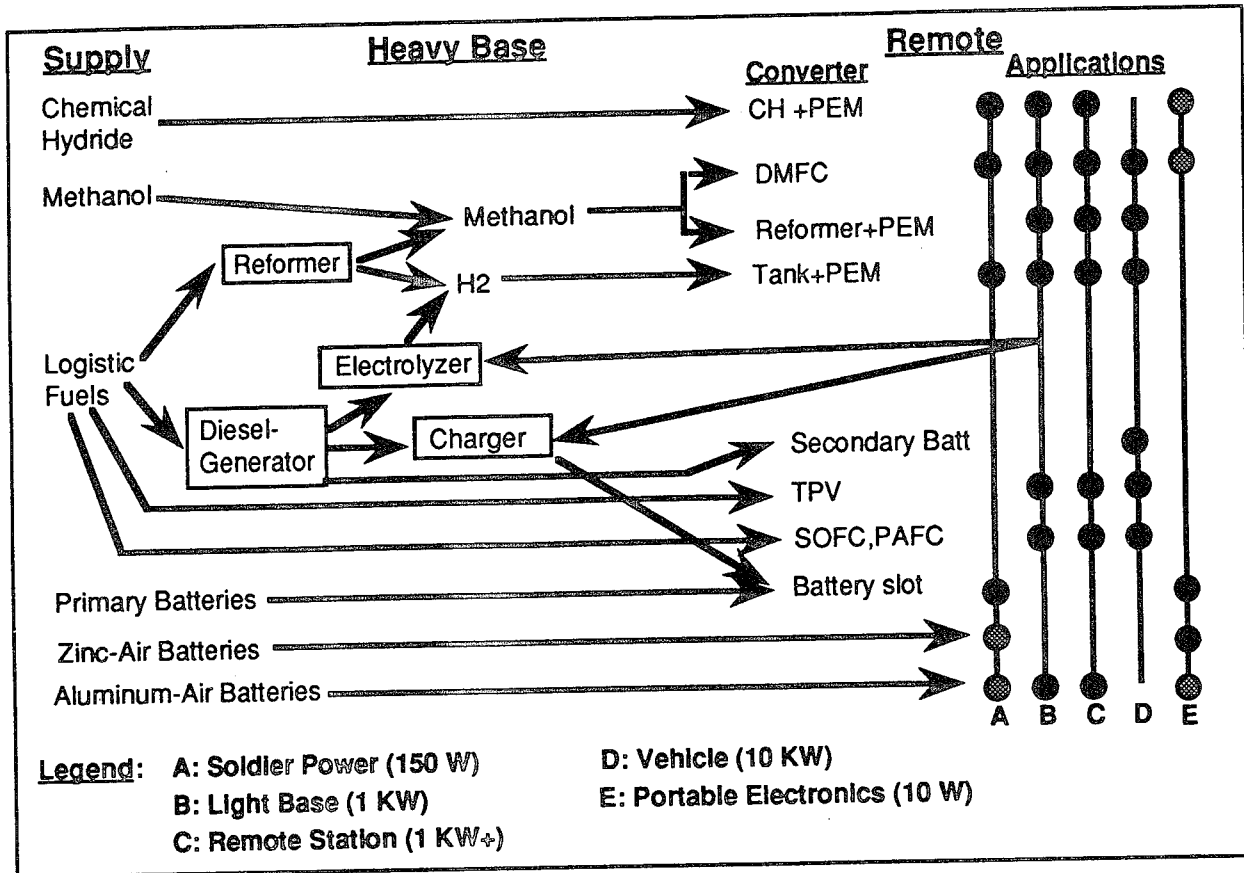


Figure 1. Portable and mobile power systems logistics grid from fuel source to remote site application

II. Attributes of Fuel Cell Systems

Fuel cell systems, in general, can be classified according to the four categories/types as follows: PEM, direct methanol PEM, PAFC, and SOFC. The advantages of the PEM fuel cell design are its low temperature operation, fast start-up characteristics, tolerance to abuse, and the fact that it represents an immobile electrolyte. There are some limitations, however, such as the low temperature limits, the requirement for a high quality fuel (purification, non-contamination constraint) for efficient operation, and the overall relative expense when compared to non-PEM alternatives. The materials required are available in large quantities if they are needed for mass production purposes.

When the direct hydrogen PEM system is evaluated in terms of its favorable and less favorable attributes, the system is considered to be environmentally friendly when carefully handled. Hydrogen is available in sufficient quantities for the task. In terms of mass and weight, the system mass/weight factor could prove to be less optimum than

desired due to the hydrogen-container or generator requirements. Most of the participants believed the safety aspect will dominate all portions of the logistics infrastructure. It is anticipated that, initially, hydrogen will be supplied from new high pressure tanks, and subsequently from the reaction of chemical hydrides with water.

When evaluating the direct methanol system, one should consider that the fuel supply system is environmentally friendly, can be readily stored, and is easily transported. Its major potential advantage is a higher energy density than many alternatives being considered (chemical hydride may be higher). The design eliminates the need for a reformer at the fuel cell, resulting in a lower weight, smaller volume, and expected reduced costs. The less favorable attributes which must be weighed include its greater use of catalysts, need to reduce methanol crossover within the system (complex technical problem when working to increase efficiency at various operating conditions), and the need for access to a ready water supply. Sealants must be developed further to extend its lifetime and operating parameters, and its relative sensitivity to orientation must be overcome to assure its best performance. It is believed that this system is approximately three years behind the hydrogen-air system, although some issues have been resolved, such as control of water generation and thermal management at higher temperatures (200 degrees Celsius). Stack development is lagging that of hydrogen-air, among others. Although applications for military and civil use in transportation are prevalent, there are outstanding questions, i.e. whether cold-start problems and a relatively slow transient response will permit general introduction into a variety of military and civil applications.

As noted by one of the speakers, there have been at least two major workshops during the past five years which have published recommendations on development of the direct methanol fuel cell. One workshop was sponsored by DOE (1990) and another by DOE and ARPA jointly (1994). The four major recommendations, which are currently being addressed in various research programs, include the following:

- Understanding why a platinum-ruthenium catalyst system works well for the methanol oxidation reaction,
- Understanding the methanol cross-over problem and developing designs or procedures to cope with or reduce that effect,
- Developing oxygen reduction catalysts that are methanol tolerant, and

- Developing reliable models of direct methanol fuel cell stack performance.

Numerous speakers presented the state-of-the-art of direct methanol fuel cell systems based upon the achievements of several contractors working on research programs sponsored by ARPA and others. It is believed that progress toward developing efficient and compact direct methanol fuel cell is very encouraging. Advances are still needed in performance of all components, in understanding fundamental design, and in cost reduction. The Group participants urge the reader of this report to scan the speakers' viewgraphs and presentation summaries for additional information. Data from speakers' viewgraph presentations are summarized in Figure 2.

State-of-the-Art in Direct Methanol FC System Performance

ARPA DIRECT METHANOL FUEL CELL PROGRAM

<u>Single Cell:</u>	0.33V @ 800 mA/cm ² 0.41 V @ 640 mA/cm ² 258 cm ² = 67 W/Cell	Life Test	<u>Single Cell:</u>	0.45 V @ 300 mA/cm ² for >200 hrs. continuous >500 hrs. intermittent
<u>4-Cell Stack:</u>	0.4 V @ 430 mA/cm ² 258 cm ² = 178 W		<u>5-Cell Stack:</u>	0.56 V @ 100 mA/cm ² (48 W) for > 400 hrs.
<u>Efficiency:</u>	25-30% @ <300 mA/lm ²			
<u>Fuel Utilization:</u>	>70% @ 300 mA/cm ²			

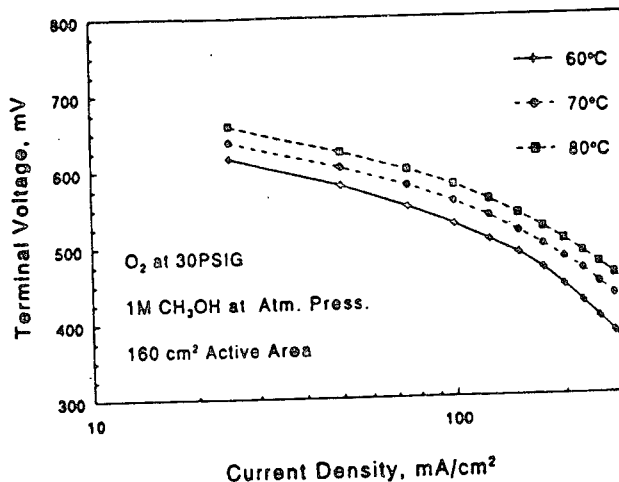


Figure 2. Effect of temperature on performance of 160 cm² Direct Liquid Methanol PEMFC (Giner, Inc. 10/93). Data summarizing the SOA of direct methanol fuel cell performance as presented at the Army Research Office-sponsored Small Fuel Cell Workshop, Prospector VII, December 3, 1994, Durham, NC

When the phosphoric acid fuel cell concept is evaluated in terms of its attributes, one notes that there are several favorable ones, including the fact that 1.5kW methanol units were constructed in 1984 for Belvoir using a reformer design. Additionally, there are 200-kW-size units being delivered to commercial customers. This concept is considered to be the most mature technology of those under examination at the Workshop for large-size units. The Group concluded by saying that the phosphoric acid FC system is relatively environmentally friendly.

The final candidate in the FC category was the solid oxide fuel cell. The Group evaluated the solid oxide fuel cell SOA and concluded that the mature development of this concept was clearly five or more years into the future. It has excellent attributes, however, in that it contains no noble metal catalyst, offers potential for operation with methanol, is carbon monoxide tolerant, and operates under ambient pressure. On the negative side, the design has a low cycle life and has a disadvantageous start-up time for the Army's portable power scenario. This system was considered not to be a near-term solution, and it was not considered as an option for the Army.

III. Attributes of Non-Fuel Cell Systems

Non-fuel cell concepts were evaluated in terms of the Army's portable power system requirements. The aluminum-air FC concept has several favorable attributes, namely an infinite shelf life, built-in safety features by virtue of the non-combustible fuel, a relatively straight-forward set of scaling laws, and a wide range of operation due to the safety aspect. There are a couple of attributes which could play either way, specifically, pursuing the option of a saline or alkaline solution as the electrolyte, and achieving in excess of 500 W-hr/kg without an electrolyte. The negative attributes include the fact that it is orientation sensitive (and requires considerable work to become orientation insensitive at a moderate cost), and is not easily recharged for turn-around operation (this is not a problem when treated logistically as a primary battery). Participants could not address the disposal or re-manufacture after life expectancy has expired due to several unknowns, such as cost, in the equation. Disposal is not a difficult problem with aluminum-air systems.

According to one of the speakers, another candidate concept in the non-FC category includes thermophotovoltaic radioisotope systems. Their attributes include no maintenance, no refueling requirements (no depot), high reliability and availability of fuels/materials for operation and mass production, no start-up or controls issues of note,

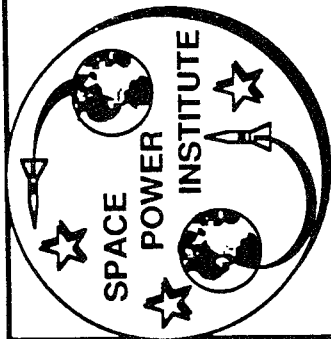
and ambient pressure operation. On the negative side, there are at least three years of development time required for TPV/radioisotope systems, and disposal costs may be prohibitive. As determined in a previous Prospector workshop, the services' general acceptance of isotope-powered systems should not be taken for granted. As an alternative to TPV/isotope powered systems, the Group examined a novel concept which coupled the TPV with an abundant logistic fuel source. The positive aspects include the fact that this system functions well at ambient pressure and has a stated potential for good conversion efficiency (after further development). Its less favorable attributes include its high temperature operation (a requirement for efficiency) and the three years needed for further research before full-scale development and testing can commence.

CONCLUSIONS

In summary, the Alternative Fuel Cell Related Technologies Working Group stated that the over-riding positive and negative attributes which tend to limit small fuel cell development could be summed up in terms of cost of manufacture, safety aspects during handling and military/civil operation, and mass/weight challenges for the truly small, portable FC systems required by the Army. It is believed that technical issues, such as energy density, cross-over, operating temperature limitations, optimal start-up characteristics and performance, and endurance (life cycle before replacement or refueling) will eventually be solved or resolved through enhanced research, improved comprehensive modeling of systems, and better materials selection and optimization.

The Group concluded its evaluation by stating there was no "best solution," because time must play a part in the decision on which FC concept should be pursued. The Group emphasized that, given the luxury of additional time, say three to five years, many of the candidate systems could have additional attributes in their favor. *As a result, the Group concluded that, in the time-phased solution, the best candidates beyond all primary and secondary batteries would be pressurized hydrogen with PEM as the first likely near-term option, followed by a chemical hydride system with PEM,* followed finally by the maturing direct methanol with PEM concept. As a further alternative, the metal-air battery concept seems to offer promise for small, portable FC power systems for both Army and civil applications under the 1kW level.*

* Sponsor's Note: Since the Workshop was held, Analytic Power Corp. has demonstrated chemical hydride fuel systems which can handle many start/stop cycles. This achievement removes one of the major obstacles to adoption of chemical hydride fuels.



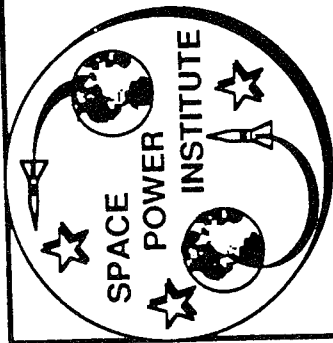
Prospector VII: Small Fuel Cells for Portable Power

Alternative Fuel Cell Related Technologies

SUMMARY BRIEFING

Group B

Washington Duke Inn, Durham, NC
October 31 - November 3, 1994



GP B PARTICIPANTS

o Robert Hamlen
Eugene Smotkin

ARL [chair]
Illinois Inst Tech [asst. chair]

o Bhaskara Rao
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Pete Howard
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Eric Simpkins
Dick Paur
Richard Weller

Explorex
Argonne lab
DoD
ARPA
w/ARPA
Indiana - Purdue
Belvoir
Natick
Royal Military
Energy Res Corp
ARO - sponsor
NSWC

Alternative Fuel Cell Related Technologies Working Group
Prospector VII: Small Fuel Cells for Portable Power

Working group 2 - Alternatives

- Identify alternatives to fuel cells in small size ranges - provide brief summary of status of each and size range of application
- Assess alternative fuel cell technologies for systems under 1 KW - summarize +'s & -'s
- Identify and characterize (prioritize) 'self-contained' fuel cell options and size range of application - notion of 'throw-away' fuel cell system - summarize SOA

It is anticipated that this WG will finish early and will split up to join the other two WG's

Applications

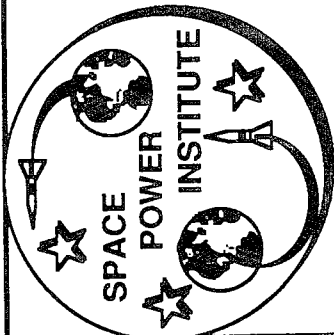
- 10W
- 150W
- 1000W

Fuel Cell Options

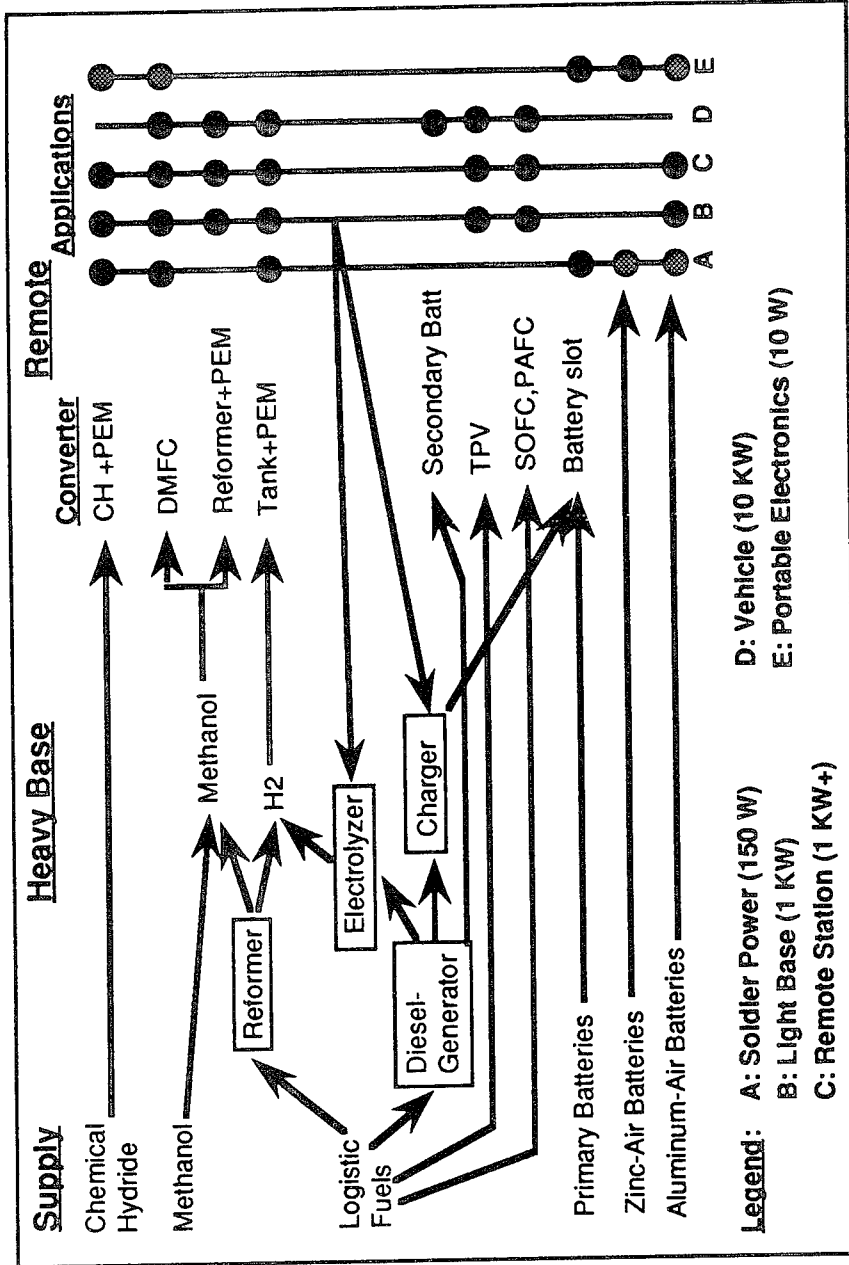
- **Hydrogen PEM**
 - Chemical Hydride
 - Pressurized Hydrogen Gas
 - Physical Hydride
 - » Lower Energy Density
- **Methanol PEM**
 - Direct Methanol PEM
 - Reformer with PEM
- **Logistic Fuel**
 - SOFC
 - PAFC

Non-Fuel Cell Options

- **Metal-Air Batteries**
 - Aluminum-Air
 - Zinc-Air
- **Thermophotovoltaics (TPV)**
- **Alkaline-Metal Thermoelectric Converter (AMTEC)**



Portable and Mobile Power Systems



**Alternative Fuel Cell Related Technologies Working Group
Prospector VII: Small Fuel Cells for Portable Power**

Group B

ATTRIBUTES OF FC SYSTEMS - 1

PEM - Environmentally-friendly

- ✓ Low temperature
- ✓ Fast start-up
- ✓ Tolerance to abuse
- ✗ Low temperature limitation
- ✗ Precious metal catalyst
- ✗ Requires high quality fuel
- ✗ Expensive, but available
- ✓ Immobile electrolyte

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Group B

ATTRIBUTES OF FC SYSTEMS - 2

PEM - Direct H2

- ✘ Environmentally OK
- ✓ Available
- ✘ Weight, volume
- ✘ Safety of H2 supply
- ✘ Logistics problems

Summary Briefing

Group B

ATTRIBUTES OF FC SYSTEMS - 3

PEM - Direct CH₃OH; environmentally OK

- ✓ **Friendly fuel**
- ✓ **Higher energy density ★★**
- ✗ **Catalysts**
- ✗ **Crossover**
- ✗ **Water supply**
- ✗ **Sealants**
- ✗ **Sensitivity to orientation ★**

At least three years behind H₂-air ★

- ✓ **Water & thermal management**
- ✓ **Logistics**
- ★ **Stack development**
- ★★ **Chemical hydride may be higher**

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Group B

ATTRIBUTES OF FC SYSTEMS - 4

PEM - Direct CH₃OH (continued)

- ✓ Dual-use in transportation, etc
- ? Cold-start problems
- ? Transient response

ATTRIBUTES OF FC SYSTEMS - 5

ix Phosphoric Acid Fuel Cell

- ✓ 1.5kW methanol units made (1984) for Belvoir - reformer
- ✓ Megawatt units under test
- ✓ Most mature technology

Environmentally OK

491

Group B

ATTRIBUTES OF FC SYSTEMS - 6

☒ SOFC - environmentally OK

✗ 5+ years in future

✓ No noble metal catalyst

✓ Potential for 500°C operation with CH₃OH

✗ Power profile specific

✓ CO tolerant

✗ Start-up time

✓ Ambient pressure operation

Group B

Single Mission Power Source - supplied from factory

 x PEM system

- Low cost membrane (available)
- Above 50W
- Chemical hydride
- Low cost catalyst
- 300-400 W-hr/kg

 x Alkaline Methanol?

 x OR Chemical Hydride?

493

Group B

Summary Briefing

Zn - air

- * Air cathode - rate limited**
- * Power profile limited**
- * One shot once activated**
- ✓ 300 - 400 W-hr/kg**
- ✓ Safe and simple**
- * Aqueous Hg - from Zn**
- * Poor low temperature operation**
- ✓ Low cost**

Disposal?

Group B

- Aluminum - air**
- ✓ **Infinite shelf-life**
- ✘ **500+ W-hr/kg without electrolyte**
- ✓ **Easy to scale**
- ✘ **Option for saline or alkaline electrolyte**
- ✓ **Safety**
- ✓ **Wide range of operation**
- ✘ **Orientation sensitive**
- ✘ **Requires work to become orientation insensitive moderate cost**
- ✘ **Not easily recharged**
- Disposal?**

495

Group B

- Thermophotovoltaic (TPV) - radioisotope**
 - ✓ **No maintenance, no refueling (no depot)**
 - ✓ **High reliability, availability**
 - ✓ **No start-up, controls issues**
 - ✓ **Ambient pressure operation**
 - ✗ **At least 3 years development**
 - ✗ **Disposal issue - cost**
 - ✗ **Service's acceptance of isotope systems**

- Thermophotovoltaic (TPV) - logistic fuel**
 - ✓ **Ambient pressure operation**
 - ✓ **Potential for good conversion efficiency**
 - ✗ **High temperature operation**
 - ✗ **At least 3 years development**

WORKING GROUP III SUMMARY

PROSPECTOR VII: SMALL FUEL CELLS FOR PORTABLE POWER

GROUP C SUMMARY

Key Research Issues Pacing Development

INTRODUCTION

The Key Research Issues Working Group was tasked with developing a consensus on the critical research impediments to be resolved for the successful, rapid development of small fuel cell power systems for the individual soldier. The workshop body was to prioritize elements within their respective topic areas in terms of the most vital systems, research issues, technologies, and options which would ensure success in the fuel cell R & D program. In the process of the discussions, the state-of-the-art in terms of performance and target parameters was to be established. *A measurable outcome of this Workshop would be the identification of the basic scientific support which the participants believe should be earmarked by research and funding planners to assure small fuel cell improvement and development.*

The general charge to the Workshop body stated as a "given" that the military will always be looking for better power sources. The current emphasis is on the Soldier System, i. e. the system which is carried by, or supports, the individual soldier in the field. For the purposes of the Workshop, participants were to assume that the entire system had to be carried by the individual soldier, thus ***the system's energy density was to be considered of paramount importance*** in establishing parameters, planning factors, limitations, and options for technology development. Furthermore, in evaluating the "small system," the definition for "small" was defined as "any system less than 1kW at the upper end." Preferably, the smaller the system, the more advantageous it would be due to the lowering of the soldier's combat load (it would thus produce an equivalently lower total system mass). Specific guidance, any constraints imposed by the sponsor, and the individual "charge to Group C," were as follows:

- Develop a consensus of the leading technical challenges and show-stoppers limiting the rapid development of small fuel cell systems. Prioritize these for the leading fuel cell technologies.
- Identify state-of-the-art performance and goals/targets for further work.
- Summarize potential strategies for short term improvement and for future "technology break throughs."
- Identify basic scientific support needed for fuel cell improvement and development.

PARTICIPANTS

The Working Group participants were drawn from military, industry, and academic sources. The Group was evenly divided, with 33% of the participants being from the military sector, 30% from industry, and the remainder (37%) being from the academic sector. The rich experience and broad background in fuel cell research and development provided a base line for fruitful discussions and meaningful recommendations on a number of key research issues. The list of participants are shown below.

- Bob Savinell, Chairman, Case Western Reserve University
- Bob Nowak, Co-Chairman, ONR
- Cal Johnson, Assistant Chairman, Auburn University
- Larry Dubois, Assistant Chairman, ARPA
- Peter Fedkiw, North Carolina State University
- Ralph White, University of South Carolina
- Michael Binder, US Army Research Lab
- Charlie Martin, Dow Chemical Company
- Oliver Murphy, Lynntech
- Michael Krumpelt, Argonne National Laboratory
- Karl Wally, Sandia Laboratories
- Dave Bloomfield, Analytic Power
- Nguyen Minh, Allied Signal
- Joe Stockel, US Gov't
- Harvey Frank, Jet Propulsion LAB
- Bruce Tatarchuk, Auburn University
- Jose Giner, Giner, Inc.
- Warren Buck, Dupont Nafion
- S. Srinivasan, Texas A & M University
- Bill Peters, MIT Energy Laboratory
- William O'Grady, NRL

DISCUSSION

The working group decided to limit its discussion to portable power requirements necessary for the individual soldier. The requirements were divided into two primary functional areas. First, microclimate cooling which requires power on the order of 150 watts and up; and second, all other electronic devices requiring power from 5 watts up to ~50 watts. As shown in Figure 1 on the next page, individual soldier power requirements may be viewed as two broad functional areas, microclimate cooling and all other system requirements. How the power package is assembled and/or the power managed may vary. The total power requirement of approximately 200W may be treated a single source or as a distributed source as shown in Figure 1.

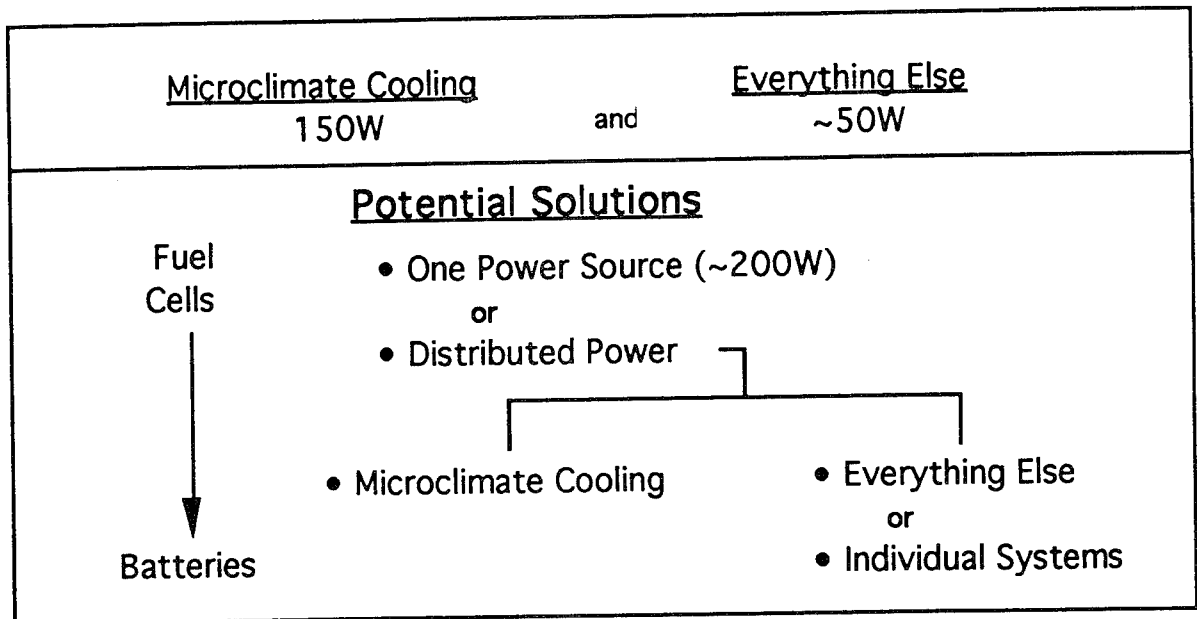


Figure 1. Individual Soldier Power Requirements and Potential Solutions

In one category, a single power source (approximately 200 watts) will provide all the energy needs for the individual soldiers. Alternatively, a distributed power system of two major types could be developed. One power system would be only for microclimate cooling, the second power system would include everything else. Furthermore, this second power system can be further divided into two modes; a single power unit that supplies all the needs, or distributed individual systems for the various components. *A single source 200 watt unit would, by necessity, probably be a fuel cell system.*

Power(W)	Energy(W-Hr)	Wt(Kg)/V(L)	Technology
25	300	1/1	Battery/Fuel Cell
150	>600 - 10,000	5/4	Fuel Cell
>150			
<u>Potential Fuel Cell Solutions</u>			
<ul style="list-style-type: none"> • He/PEM • MeOH/PEM • MeOH/SOFC 			

Figure 2. Power/Energy Targets for Future Work and Potential Solutions

On the one hand, as further depicted in Figure 1, sub-dividing power source into a distributed system of various sizes could result in some systems capable of being handled by batteries. So the range of power needs could be fulfilled by fuel cells or batteries, depending on the specific mission. As shown in Figure 2, it is envisioned that a 50 watt power system will be met by two devices of 25 watts each. The energy requirement is estimated to be ~300 watt hours requiring less than 1 kg mass and 1 liter of volume. *This energy need possibly could be met by fuel cells or by batteries.* The ultimate driver in each option will be system energy density.

On the other hand, the 150 watts and above power source will require energy levels on the order of 600 up to ~10,000 watt-hrs. It will need to weigh less than 5 kg and have a volume less than 4 liters. *This particular need will certainly require a fuel cell.* The working group decided that the fuel cell technologies which are available at this time and are most amenable for these applications include the hydrogen PEM, the methanol PEM and the methanol solid oxide fuel cell (SOFC). As a result, all research issues discussed revolved around these three FC systems.

There are a number of generic research issues which must be addressed to develop these technologies to the point of being useful in the field. Group C chose to focus its discussion on several **primary research-related issues** which are discussed in greater detail in the following sections.

Primary Research Issues

Fuels and Fuel Storage - This issue includes the need for strategic decisions on what fuels will be and should be used in the field and how these fuels should be stored. It was the feeling of the workshop participants that the two most likely candidates for fuels include hydrogen and methanol. The specific problems identified primarily dealt with the storage of hydrogen, either in chemical hydride form or in high pressure containers. It was felt that the storage of methanol is not a particular problem at this time. Several solutions were suggested for addressing the issue of reaction kinetics control and load following ability of hydride storage. These solutions include: optimization of composition morphology and advanced reactor engineering. It was also suggested that potential solutions to the storage problem may be obtained by looking for innovative hydrogen production chemistry which can be used in the field for on-site hydrogen generation.

Stack Design and Materials - This issue deals with the need to reduce the weight, size and cost of the various components of the fuel cell stack. Ultimately, it will be necessary to increase stack power density. Solutions will include making thinner cell repeat units using new, lighter and lower cost materials. Stack designs to decouple heat and reaction flow must be developed and in particular, novel approaches must be used for developing alternative stacking system designs. This point emphasizes the fact that it may be necessary to abandon the traditional type of fuel cell design approaches and to look for novel approaches that can significantly reduce the volume and complexity of a fuel cell stack. For solid oxide fuel cells, a

need exists for defect free and tough ceramics. These materials must be able to have proper thermo-expansion coefficients to adjust for rapid start and rapid cool down conditions. All fuel cell systems have issues relating to safety and control which must be addressed.

Electrolytes - There were a number of specific problems, some of which are generic, which were identified relevant to this issue. The working group made an attempt to categorize the specific problems and to identify whether they are relevant for the PEM and/or DMFC and/or SOFC systems. Figure 3 depicts this relationship of FC problems to three specific categories of systems.

<u>Specific Research Problems</u>	<u>PEM</u>	<u>DMFC</u>	<u>SOFC</u>
• Limit water transport (elec drag)	X		
• Water vapor pressure suppression	X		
• Increase conductivity w/good mech prop	X	X	X
• Increase dimensional stability	X	X	X
• High/low temperature operation	X	X	
• Reduce MeOH crossover		X	
• Stability in oxidizing and reducing environ			X

Figure 3. Electrolytes - showing relevancy for various FC systems

A number of short term and long term solutions were presented as a result of the list of electrolyte-related research issues identified by the Group. For example, in the short term, modifying presently available membrane materials by using additives, making composites, varying thicknesses, modifying equivalent weights, and altering ionic strengths were suggested as possible means of overcoming some of the specific electrolyte-related problems. Long term solutions include developing a true proton conductor. The idea of modifying membranes with dense phase films was also thought to be a long term solution for eliminating the methanol cross-over problem. On the topic of solid oxide fuel cells, there is a need to develop new oxide conductors for low temperature operation. In all electrolyte types, better theoretical and experimental understanding of conductivity, mechanisms, transport processes and physical properties are needed.

Catalysis - Again, a number of catalytic issues were identified, some related to PEM, some related to DMFCs and some related to SOFCs. Several of these issues relate to more than one of these systems and are described in Figure 4 which follows.

<u>Specific Research Problems</u>	<u>PEM</u>	<u>DMFC</u>	<u>SOFC</u>
• Slow MeOH oxidation kinetics at low temp		X	
• Slow oxygen reduction kinetics	X	X	
• Catalyst poisoning (fuel and air)	X	X	
• MeOH tolerant cathode		X	
• Need catalysts for SOFC			X
• Synthesis and characterization techniques are inadequate	X	X	X

Figure 4. Catalysis - showing relevancy for various FC systems

In general, slow methanol oxidation kinetics and slow oxygen reduction kinetics at low temperatures are still problems which must be addressed. In the DMFC, there is still a need for an active cathode catalyst which is methanol tolerant. Catalyst poisoning from various sources in both the fuel and air side of the fuel cell must be addressed. This includes contaminated air, processed fuels with sulfur and other tenacious poisons and perhaps even the issue of contamination as a result of mission activity. In general, it was felt that there is a need for better synthesis and characterization techniques to develop reproducible well-understood catalysts. Solutions were identified for short term and long term implementation. In the short term, one important task will be to identify tests for potential poisons in fuel cell operation. A number of specific materials and catalyst studies were also suggested. In the long term, catalysis issues would benefit from implementation and development of micro-kinetic modeling approaches and the development and application of new experimental methods to elucidate reaction mechanisms.

System Engineering/Modeling for Fuel Cells - In order to develop, optimize, scale-up and match performance to mission requirements, mathematical models of fuel cells must be developed. Specific problem areas include thermo and water management models. Reactant distributions are difficult to model and algorithms must be developed for this purpose. In addition, models are not available for predicting start-up and transient effects. The working group suggested that models be developed at a number of levels. Molecular levels are needed for predicting properties and kinetic mechanisms as well as for relating molecular and morphological properties to macroscopic transport physical properties. An effort is needed for detailed models which describe reactant flow and thermo-transport at the cell and the stack level. It is also felt that the integration of models at all levels will be critical for understanding, designing, and optimizing system performance. This may require developing and implementing advanced mathematical treatments for very non-linear and very stiff, large systems of equations.

Some concluding remarks were generated to represent the Group's findings and recommendations regarding the impact of key research issues upon the development of small fuel cells for the individual soldier.

SUMMARY

First, the Working Group recognized that the development of fuel cell systems of 25 watts to less than 150 watts for military applications could lead to numerous commercial applications as well as military applications. Although military applications were the primary motivator for this particular workshop, one should seriously consider the commercial spin-offs due to the vastly increased market and investment opportunity. Consequently, it was the opinion of the Group that an emphasis on research to resolve these fuel cell technical issues will benefit civilian markets as well as military needs, and this "dual market opportunity" should be pursued.

The Group fully concurred with the General Charge made by the sponsor, in that, indeed, the system's energy density is of paramount importance in establishing parameters, planning factors, limitations, markets, and options for technology development. Furthermore, in evaluating the "small system," the definition for "small" was defined as "any system less than 1kW at the upper end." The Group also agreed with the premise that, the smaller the system (the total available, but sufficient, energy), the more advantageous it would be due to the lowering of the soldier's combat load (it should produce an equivalently lower total system mass).

Finally, the Group determined that the most relevant key research issues, distributed into five major categories, should be the selection and use of fuels and fuel storage methods, stack design and materials, electrolytes, catalysis, and systems engineering /innovative modeling techniques for fuel cells.

At the conclusion of Working Group deliberations, the Co-Chairman presented the Group's findings to the collective Workshop for approval and comment. The main briefing points were well-received. The Group's briefing slides are included in the Proceedings.

**Prospector VII:
Small Fuel Cells for Portable Power**

KEY RESEARCH ISSUES

Group 3

**Washington Duke Inn
Durham, North Carolina
October 31 - November 3, 1994**

PARTICIPANTS

Working Group 3

- Bob Savinell, Chairman & Guru, Case Western Reserve University
- Bob Nowak, Messenger, (Hold your fire!!), ONR
- Cal Johnson, Assistant Director, Auburn University
- Larry Dubois, Kibbitzer, ARPA

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Ralph White, University of South Carolina

Michael Binder, US Army Research Lab

Charlie Martin, Dow Chemical Company

Oliver Murphy, Lynntech

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Karl Wally, Sandia Laboratories

Dave Bloomfield, Analytic Power

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Joe Stockel, US Gov't

Harvey Frank, Jet Propulsion LAB

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Jose Giner, Giner, Inc.

Warren Buck, Dupont Nafion

S. Srinivasan, Texas A & M University

Bill Peters, MIT Energy Laboratory

William O'Grady, NRL

GENERIC RESEARCH ISSUES

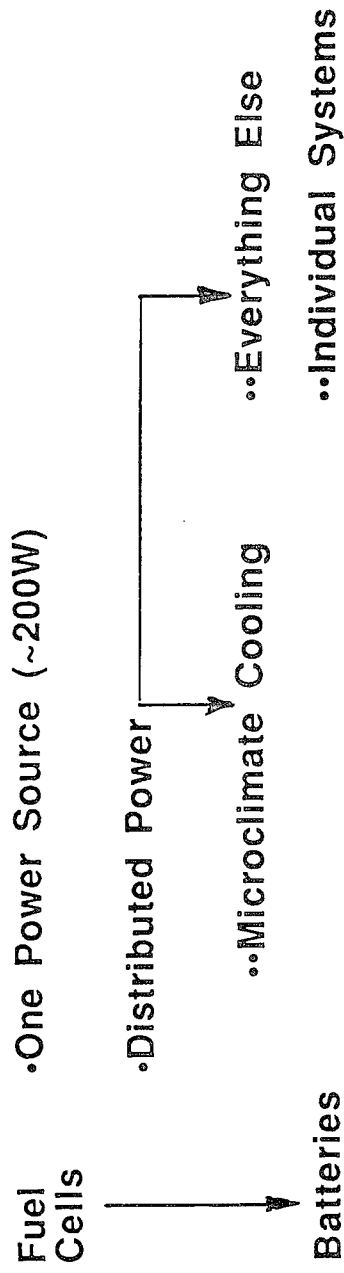
- Everything Else!
- Fuels & Fuel Storage
- Stack Design and Materials
- Electrolytes
- Catalysis
- System Engineering/Modeling
- Everything Else!!

INDIVIDUAL SOLDIER

POWER REQUIREMENTS

Microclimate Cooling Everything Else
~150W ~50W

Potential Solutions



Issue: Fuels & Fuel Storage

Specific Problems:

- Chemical hydrides
 - Controlling reaction kinetics/load following
 - Suppressing hydrate formation
- High pressure hydrogen
 - Small, lightweight tankage
 - Safety concerns

Solution(s):

- Optimize composition and morphology of chemical hydrides
- Reactor engineering (chemical, thermal)
- Innovative hydrogen production chemistry

Issue: Stack Design and Materials

Specific Problems:

- Weight/size/cost of bipolar plates, flow fields, and end plates
- Increase stack power density
- Need defect free and tough ceramics for rapid start (SOFC)
- Safety and control

Solution(s):

- Increase cell pitch (thinner cells)
- Lighter weight/lower cost materials
- Stack redesign to de-couple heat and reactant flow
- Match thermal expansion coefficient (SOFC)
- Novel approaches - mixed reactant systems, alternate stack/system designs

Issue: Electrolytes

Specific Problems:

	PEM	DMFC	SOFC
• Limit water transport (electroosmotic drag)	X		
• Water vapor pressure suppression	X		
• Increase conductivity w/good mech prop	X	X	X
• Increase dimensional stability	X	X	X
• High/low temperature operation	X	X	
• Reduce MeOH crossover		X	
• Stability in oxidizing and reducing environments			X

Solution(s):

- Short Term:
 - Membrane modifications such as additives, composites, varying thicknesses, equivalent weight, ionic strength
- Long Term:
 - Develop "true" proton conductors
 - Modify membranes with dense phase films (e.g. Pd)
 - Develop new oxide conductors for low temp operation
 - Better theoretical and experimental understanding

Issue: Catalysis

Specific Problems:

	PEM	DMFC	SOFC
• Slow methanol oxidation kinetics at low temp		X	
• Slow oxygen reduction kinetics	X	X	
• Catalyst poisoning (fuel and air)	X	X	
• Methanol tolerant cathode		X	X
• Need catalysts for SOFC			X
• Synthesis and characterization techniques are inadequate	X	X	X

Solution(s):

- Short Term:
 - Identify and test potential poisons
 - Develop intermetallics, metal/metal oxides, additives/promoters, and alloy catalysts (e.g. optimize Ru/Pt for DMFC)
 - Test crystal size and orientation effects
 - Macrocycles & oxometallates
 - Electrocatalysts at 450C for SOFCs
- Long Term:
 - Microkinetic modeling approaches
 - New experimental methods to elucidate mechanisms

Issue: System Engineering/Modeling of Fuel Cells

Specific Problems:

- Thermal and water management models are limited in scope or are system specific
- Reactant distributions difficult to model
- Start-up and transients modeling not available

Solution(s):

- Develop molecular-level models in the cell
- Develop detailed reactant flow and thermal models at all cell and stack levels
- Integration of models (at all levels) is critical to understanding, designing, and optimizing system performance

POWER/ENERGY TARGETS FOR FUTURE WORK

<u>Power, W</u>	<u>Energy, W-Hr</u>	<u>Wt, Kg/V.l</u>	<u>Technology</u>
25	300	1/1	Battery/Fuel Cell
150	>600-10,000	5/4	Fuel Cell

>150

Potential Solutions

H₂/PEM

MeOH/PEM

MeOH/SOFC

APPENDICES

APPENDIX A

APPENDIX A

NSF/DOE FUEL CELLS FOR ELECTRIC AUTOMOBILES WORKSHOP

Washington, DC
December 14-16, 1993

Workshop Chairman: Vernon Roan
University of Florida

Attachment: Working Group Summary entitled "Fuel Cell Membrane and Electrochemistry Research Issues"

Working Group Chairman: Supramaniam Srinivasan
Secretary: John Kosek

The Prospector VII participants and sponsors wish to acknowledge the contributions of members of a prior Workshop, the NSF/DOE-sponsored Workshop on Fuel Cells for Electric Automobiles, held in Washington, DC, December 14-16, 1993. The attached summary of discussions from the Working Group on "Fuel Cell Membrane and Electrochemistry Research Issues" proved useful in assisting the members of Prospector VII's Key Research Issues Working Group as they reviewed and prioritized issues in the electrochemistry and fuel membrane categories. *That summary of discussions from the December, 1993, meeting is hereby included as Appendix A to complete the Prospector VII Proceedings.*

**Report of Working Group on "Fuel Cell Membrane
and Electrochemistry Research Issues"**

Chairman: Supramaniam Srinivasan

Secretary: John Kosek

1. Participants and Affiliations

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Ben Tefertiller, Dow Chemical Company
Bill Wepfer, Georgia Tech

2. Objectives

The objectives of this working group were to (i) assess the status of the technology of fuel cells for electric vehicles; and (ii) identify and prioritize the research issues, in these and future fuel cell technologies, in order to meet their performance and cost goals for the electric vehicle application.

3. A Synopsis of the Current Status of Fuel Cell Technologies for Electric Vehicle Applications With Emphasis on Proton Exchange Membrane Fuel Cells

The "Fuel Cells for Transportation Applications" Program was initiated by the U.S. Department of Energy in 1977. In the initial phase of the study from 1977 to 1980, techno-economic assessments were carried out to identify the most promising types of fuel cells and fuels for this application. These studies led to the conclusions that:

(i) for the light duty, passenger vehicle application, the proton exchange membrane fuel cell (PEMFC), with methanol, reformed to hydrogen on board the vehicle, is the most logical choice; and

(ii) for the heavy duty vehicles (buses, trucks, trains, ships, etc.), the phosphoric acid fuel cell (PAFC) power plant, again with methanol reformed on board the vehicle, is the most promising system. It was also concluded that, for this application, in the longer term the solid oxide fuel cell system could play some role because of the prospects for direct utilization of methanol and other organic fuels.

In the 1980s, the research and development activities were focused in these two areas, i.e., PEMFCs for automobiles and PAFCs for buses, trucks, etc. Another interesting conclusion from the studies in the 1970s and 1980s was that because of the high power density requirements for start-up and acceleration and the high energy density requirements for the range of the vehicle, a hybrid power source, fuel cell/battery, will be the most appropriate, as evident from the Ragone Plot for energy conversion and storage systems (Fig. 1).

It is interesting to compare performances of the different types of the state-of-the-art fuel cells (Fig. 2). This figure shows that for the low temperature fuel cells utilizing hydrogen, the efficiency and power density losses are due to electrocatalytic (as seen from the initial semi-exponential region of the cell potential vs. current density plot) and ohmic and limitations (slope of the linear region). The relatively poor performance of the direct methanol fuel cell is due to the very slow kinetics of both methanol oxidation and the oxygen reduction in the fuel cell whereas in the fuel cells, with hydrogen being consumed in the anode, the electrode kinetics is slow only for the oxygen reduction reaction. In the case of the higher temperature systems (with molten carbonate or solid oxide electrolytes) the electrode kinetics of the fuel cell reactions are fast but the ohmic and mass transport losses are high.

Fig. 2 also confirms that, at the present time, the most promising fuel cell systems for the transportation applications are:

- PEMFCs utilizing hydrogen light duty for the passenger vehicles
- PAFCs utilizing hydrogen for the heavy duty vehicles

In order for the other types of fuel cells to reach the performance levels required for these applications, it is necessary to significantly increase

- (i) the electrode kinetics of the methanol oxidation reaction ; and
- (ii) considerably lower the ohmic and mass transport losses in the high temperature system.

Another factor, which has to be taken into consideration for the transportation application is the pseudo-instantaneous start-up time, needed for the passenger vehicle. With the low temperature fuel cells (PEMFCs, alkaline fuel cells-AFCs), this is possible and is particularly so with fuel cell/battery hybrid power sources. This is not the case with the high temperature fuel cells (MCFC and SOFC) at the present time. Even the PAFC which operates at an intermediate temperature, the start-up time is relatively long (a few hours).

4. Performance Issues Related to Cost-Goals of \$30-40/kW, Weight, Volume and Start-Up Time Restrictions of Power Plants

One of the toughest challenges in developing fuel cell power sources for electric vehicles is to meet the U.S. Department of Energy's goal of \$30-40/kW for the capital cost of the power plants. This target cost is necessary for the fuel cell powered vehicle to be economically competitive with the state-of-the-art internal combustion or diesel engine powered vehicle. The costs of power plants for the latter type of vehicles are of the orders of \$15-20/kW. However, one must take into consideration that it has taken nearly 100 years to advance these technologies to reach the present level of performance and costs.

In order to meet the above mentioned cost-goals, the performance issues are as follows:

- Need for a quantum jump in performance.
- Necessary to find low-cost materials and fabrication processes.
- R&D activities need to be conducted in the framework of Systems-Performance-Cost Analysis

The capital cost of the power plant is inversely proportional to the power density. The higher the power density, the higher is also the rate of heat generation and the more expensive could be the auxiliaries needed for heat rejection. This is one of the reasons for the need for R&D activities on the multicell stack to be well coordinated with the Systems-Performance-Cost Analysis.

Low cost materials for the components of the electrochemical multicell stacks, other sub-systems such as the fuel processor, and the auxiliaries (for thermal and water management, power electronics) are vitally necessary to meet the capital costs. Even in the large scale automated manufacture of a variety of products, the capital costs are about two to four times the costs of the materials. This fact shows the importance of minimizing the fabrication costs of electrodes, electrolyte layers, bipolar plates, etc. At the present time, even though the material costs are not high, the fabrication costs of the components of the multicell stack are prohibitive.

The performance issues, related to weight and volume restrictions of power plants, which are also governed by the targets set by the internal combustion and diesel engine powered vehicles, are

- Need for high power density
- Need for high energy density

The U.S. Department of Energy's goals for battery powered vehicles are 400 W/kg for the power density and 200 Wh/kg for the energy density. To approach these goals in the short to medium term, development of a hybrid power source - fuel cell/battery - is the most promising solution, but the long term solution will be to use a sole fuel cell power source. This again requires a quantum jump in power density is necessary.

The methanol fuel processor, carried on board the vehicle, is responsible for 30 to 40% of the weight and volume of the fuel cell power plant. The development of a direct methanol fuel cell with the desired energy efficiencies and power densities could thus significantly reduce the weight and volume of the reformer.

As stated in Section 3, pseudo-instant start-up times are vitally important for passenger vehicles. Though this does not appear to be a problem for low temperature fuel cells and fuel cell/battery hybrid power sources, methods to minimize the start-up times of the higher temperature systems (PAFC, SOFC) need to be developed.

5. Research Issues, Research Priorities and Recommendations

5.1 Proton Exchange Membrane Fuel Cells

5.1.1 Research Issues and Research Priorities

At the present time, there is great stimulation and enthusiasm for the development of PEMFCs for electric vehicles in several university, government and industrial laboratories in the USA and several foreign countries (Canada, Japan, France, Germany, Korea, Belgium, Netherlands, India, etc.). From a technology development point of view, the Ballard Power Systems, Inc. have made the most significant progress, as evident from their demonstrations of the 5 kW power plants and of the fuel cell powered bus.

According to the views expressed by members of the working group, the Research Issues, which were stressed, are as follows:

- Proton Conducting Membranes
 - High Temperature Capability*
 - Methanol Rejecting Membranes*
 - Water and Thermal Management
 - Modeling of Proton Transport at a Molecular Level
 - Investigations of Metal-Proton Conducting Membrane Interface
 - Stack Heat Rejection

- Electrocatalysts for PEMFCs
 - CO Tolerant Anode Electrocatalysts*
 - Direct Methanol Oxidation Electrocatalysts*
 - Alternatives to Platinum
 - Electrode Structure Reproducibility

5.1.2 Rationale and Recommendations - Proton Conducting Membranes

The development of a proton conducting membrane with a high temperature capability (about 150 to 200°C) is a "high priority" area of research because at these operating temperatures it will be possible to minimize the problems of

- (i) poisoning of the anode electrocatalyst by small amounts (say 1 to 2%) of carbon monoxide;
- (ii) low electrocatalytic activity of the oxygen reduction and methanol oxidation reactions; and
- (iii) waste-heat rejection.

The state-of-the-art proton conducting membranes contain perfluorinated sulfonic acid chains and have a Teflon like structure. The conductivity is dependent on the maximum humidification with water and on the CF₂ and SO₃H contents. Higher the SO₃H content, better is the conductivity, but at too high a SO₃H content the mechanical integrity of the membrane could be affected. The development of the higher temperature membrane, without the need for external humidification, will greatly simplify the operation of the PEMFC. Finding alternatives to the perfluorinated sulfonic acid membranes, with the desired proton conducting characteristics at the elevated temperatures, could significantly advance the PEMFC technology.

In order to develop PEMFCs using the methanol fuel directly, it is vitally important to minimize the transport of methanol from the anode to the cathode. The cross-over of methanol

* High priority research areas for short to medium term

causes a loss in energy efficiency and power density of the fuel cell. Finding a solution to this problem is also a "high priority" area of research because, elimination of the fuel processing sub-system can greatly reduce (by as much as 30-50%) the weight, volume and cost of the fuel cell power plant. The cross-over problem can probably be minimized by use of laminated or composite membranes.

The water management problem (i.e., maintaining the membrane in the completely humidified condition) is a major challenge. In practically all the PEMFC development work, the reactant gases are humidified. However, BCS Technology, a small business in the State of Texas, has developed a novel method for the preparation of the membrane and electrode assembly, for a PEMFC, which is capable of operation without external humidification and still exhibits a high level of performance (Fig. 3). More work in this direction is needed. Thermal management becomes more difficult when fuel cells are required to operate at high power densities, as in the case of the electric vehicle application. The problem is even more complex when the fuel cells operate at low temperatures, such as is the case of the state-of-the-art PEMFC. This problem can, however, be significantly reduced if higher temperature proton conducting membranes are developed. Novel methods for heat rejection, such as evaporative cooling, may be coupled with humidification of the reactant gases to solve the water and thermal management problems.

At a fundamental level, it is essential to investigate the proton conduction mechanism in the membranes. The perfluorinated sulfonic acid has an equivalent conductivity close to 1N sulfuric acid in which case proton conduction is by a hopping mechanism (Grotthus mechanism). In an aqueous electrolyte, both the anions and cations are mobile. In the case of the perfluorinated sulfonic acid, the sulfonic acid group is anchored to a side chain in the polymer and is hence immobilized. NMR and other techniques may be valuable in elucidating the proton conduction mechanism.

A related fundamental area of research, which needs to be carried out, is the investigation of the structure of the double layer at the metal/proton conducting interface. Highly sophisticated double layer studies have been conducted at metal/aqueous electrolyte interfaces and to a lesser extent at the metal/molten electrolyte interfaces, using mainly AC impedance techniques. This has not been the case at the metal/proton conducting membrane interface.

The stack-heat rejection problem is related to the thermal management problem. The major portion of the heat generated is due to the irreversible losses due to activation overpotential at the oxygen electrode. However, there are also significant contributions due to ohmic resistances, mainly of the electrolyte, and mass transport overpotential when air is used as the reactant. This research issue becomes more problematic if the future PEMFCs operate at high power densities, which will be necessary to meet the goals of low weights, volumes and costs.

5.1.3 Rationale and Recommendations - Electrocatalysts for PEMFCs

The two areas of research which have high priority and are related are (i) CO Tolerant Anode Electrocatalysts; and (ii) Direct Methanol Oxidation Electrocatalysts. Finding CO tolerant catalysts is of vital importance, since, hydrogen produced by steam reforming/shift-conversion/selective oxidation results in a small amount of carbon monoxide which causes poisoning of the platinum anode electrocatalyst. Further, even with simulated steam reformed fuels (70% H₂, 30% CO₂), the shift conversion reaction reaches an equilibrium and hence results in the production of a small amount of carbon monoxide which causes a poisoning problem, particularly at higher current density. Small amounts of carbon monoxide can also be produced even if "pristine" hydrogen is the anodic reactant because the oxygen in the air contains about 340 ppm of CO₂ and small amounts of it are transported from the cathode to the anode. Thus, by use of a dual function electrocatalyst to electrochemically oxidize the fuel and small amounts of carbon monoxide, as shown to be possible by using Pt-RuOx electrocatalysts.

One of the most challenging problems in developing DMFCs is to find electrocatalysts with high activities for the electro-oxidation of methanol. The methanol oxidation reaction is a six-electron transfer reaction and could involve formaldehyde and formic acid type species being formed as intermediates or in parallel reactions. Besides this, the adsorbed CO or COH type species cause a degradation in performance of methanol oxidation. Thus, as in the case of CO tolerant anode electrocatalysts, dual function electrocatalysts are needed to remove the poisoning species and enhance the oxidation rates of the methanol oxidation reactants.

Of a lesser priority but more of a fundamental and long range research issue is to find alternatives to platinum electrocatalyst. To date, platinum has been shown to be the most effective electrocatalyst for the hydrogen oxidation and oxygen reduction reaction on PEMFCs and PAFCs. Even though demonstrations of reductions in platinum loadings to levels of 100 µg/cm² have been demonstrated by investigators at Los Alamos National Laboratory and Texas A&M University, it is still necessary to find alternatives to platinum if PEMFCs will play a major role as power sources for electric vehicles. The reason for this conclusion is that even at this loading the Pt equivalent will be about ten times that used in the present catalytic converters and the latter application uses one third of the platinum produced each year. Thus, even for 1 million PEMFC powered automobiles manufactured per year, the platinum consumption for the automobile application will exceed that of today's platinum requirements. A basic question is why is platinum unique in exhibiting considerably higher electrocatalytic activities than others evaluated to date. Theoretical and experimental research areas are still open to resolve this question and to tailor-make electrocatalysts for future cells.

Some members of the working group made comments to the effect that "electrode structure reproducibility" is a research issue because there still exists a problem of PEMFC performance reproducibility. There is no doubt that the structure of the electrode plays an important role particularly in the efforts to reach high power density. The state-of-the-art electrodes contain substrate, diffusion and electrocatalyst layers. To lower platinum requirements and to minimize activation, mass transport and ohmic overpotentials, it is essential to optimize the structure of the electrodes and carry out detailed modeling analysis on the effect of structure of the electrodes on the performance of fuel cells.

5.2 Solid Oxide Fuel Cells

5.2.1 Research Issues and Research Priorities

The United States is in the forefront of SOFC R&D as demonstrated by the progress in this technology by Westinghouse Electric Corporation. There are also active programs in Japan, Germany and Netherlands. The SOFC R&D programs are focused on the electric utility power generating application. The advances in the R&D, particularly with respect to alternate designs, thin film structures and prospects for lower operating temperature have stimulated interests in examining this technology for electric vehicle applications.

The working group sessions led to the following Research Issues and Research Priorities (indicated by asterisks).

- Materials Studies for Interconnects and Seals*
- Lower Temperature Systems and Thin Film Fabrication Techniques*
- Basic Electrochemical Studies
 - Electrocatalysis
 - Structure of Electrode/Electrolyte Interfaces
- Thermal Cycling
- Lower Temperature Operation With Alternate Fuels

5.2.2 Rationale for Recommendations

The state-of-the-art SOFCs use (i) yttria stabilized zirconia for the electrolyte; (ii) a nickel-zirconia cement for the anode; (iii) strontium doped lanthanum manganate for the cathode; and (iv) magnesium doped lanthanum chromite for the interconnection. The tubular structure of SOFCs is the most advanced though there are efforts to develop the planar and monolithic structures.

The high priority areas of research are indicated in the preceding section. In order for SOFCs to meet the challenges, in respect to performance and costs, as power sources for electric vehicles, the following accomplishments in the R&D are vital

(i) finding electrodes, electrolytes for lower temperature operation;

(ii) developing fabrication techniques such as plasma deposition, sputtering, vapor deposition, etc. which can produce cost-effective thin film structures;

(iii) finding sealant materials through solid state science is in a very advanced state. New materials and processes have been developed in finding solids with high electronic or ionic conductivities and thin film structures have been produced. Fundamental studies to unravel the structures of the electrode/solid electrolyte interfaces, the electrode kinetics of fuel cell reactions, and the transport properties of electrons and ions in the electrode and in the electrolyte are, however, sparse. There is still a need to elucidate the contributions of activation, mass transport and ohmic overpotentials in SOFCs. Though the methods to obtain these parameters for aqueous systems are well developed, this is not the case in systems with solid electrolytes.

At the present time, SOFCs appear to have better prospects for the heavy duty applications because of the high operating temperatures. In these cases, there may not be much of a problem of maintaining the fuel cells at the high operating temperature for long periods of time. For the light duty application, it may be necessary to subject the power plants to thermal cycling. Investigations are thus necessary to determine the extents of mechanical and physico-chemical degradation caused by thermal cycling.

An advantage of SOFCs could be that internal reforming of fuels could be carried out because water is produced at the anode with the oxide ion conductors and this water could be used as the reactant for the steam reforming reaction. Further, because the fuel cell reaction is exothermic and the steam reforming reaction is endothermic and the efficiency losses for electricity production in fuel cells produce heat, a partial solution to the thermal management problem can be made by integration of the fuel cell and the reformer, as in the case of the informal reforming fuel cell. However, for the successful development of such a system with alternate fuels such as methanol or natural gas, it is still necessary to find materials for components of SOFCs for lower temperature operation (400 to 700°C rather than 1000°C as at present).

5.3 Alternate Fuel Cell Designs

The longer term Research Issues and Research Priorities will have to focus on quantum leaps in fuel cell technologies with respects to novel materials, processes, and designs. A radical change from the conventional approach is necessary to improve the performance and lower the

cost, as is necessary for the transportation applications. The research issues, which were suggested by the Working Group are

- Systems Oriented Studies to Examine Prospects of Meeting Projected Cost Goals of \$30-40/kW
- Operate PAFCs With Pure H₂
- O₂ Enrichment for Improved Performance
- Anode Exhaust Utilization of Natural Gas and Other Carbonaceous Fuels

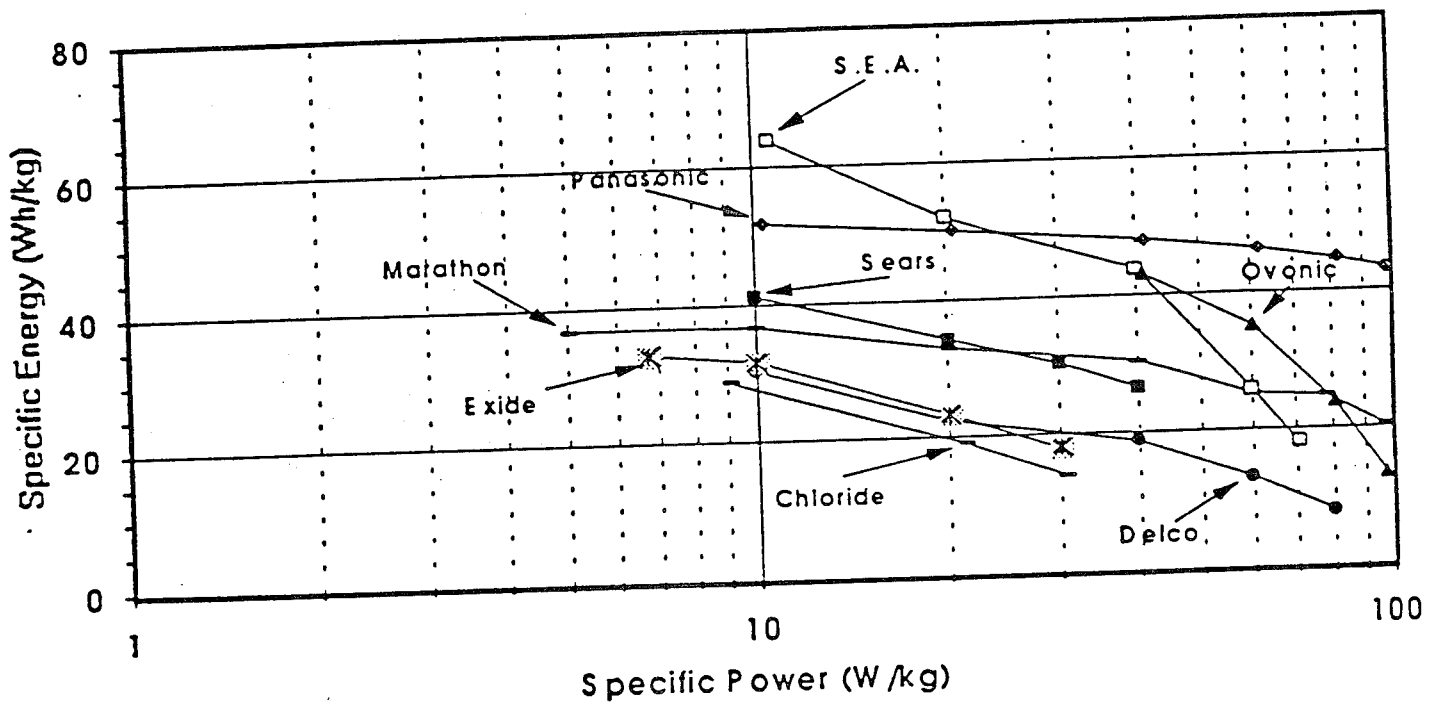


Figure 1 Ragone Plot showing the energy and power density characteristics of some commercially available battery systems. Lead Acid batteries from Chloride, Delco, Exide and Sears; Nickel-Cadmium from Marathon; Nickel-Metal Hydride from Ovonic, Panasonic and Zinc Bromine from S.E.A.

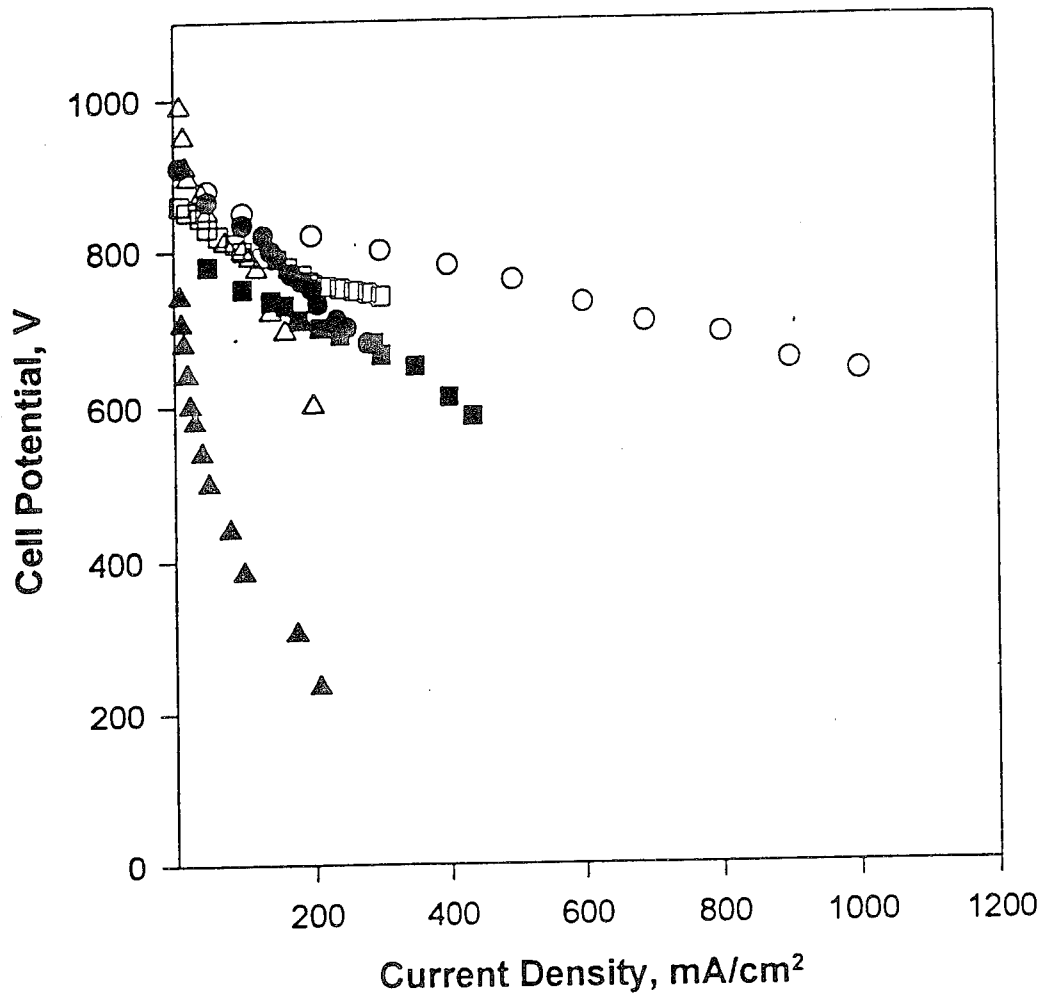


Figure 2 Comparison of performance characteristics of several types of fuel cells; (O) PEMFC, (●) MCFC, (□) PAFC, (■) SOFC, (Δ) AFC and (Δ) Direct Methanol PAFC.

Cell#-40, Hari Dhar's M&E Assembly
No humidification on both side

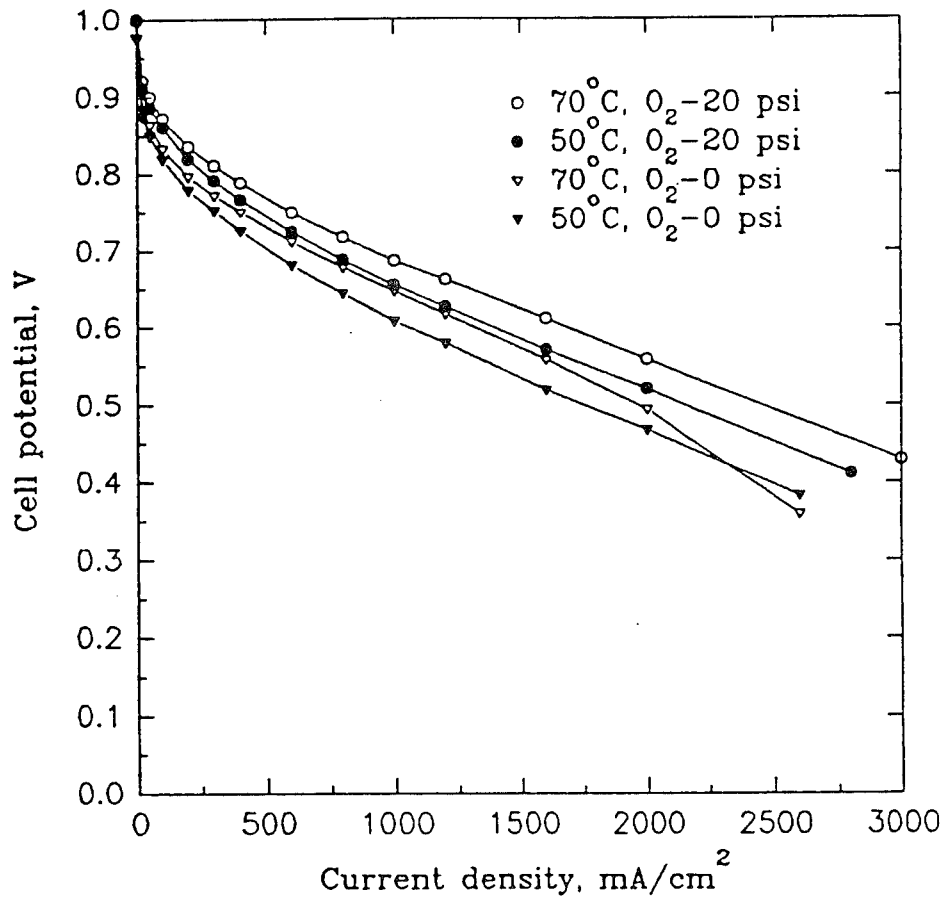


Figure 3 A plot of PEM fuel cell performance without humidification.

APPENDIX B

APPENDIX B

DOE/ARPA REVIEW MEETING

Baltimore, MD
April 26 - 27, 1994

Subject: Fuel Cells for Direct Methanol Oxidation

Introductory Remarks: Albert R. Landgrebe
Department of Energy

and

Discussion Leader: Shimshon Gottesfeld
Los Alamos National Laboratory

The Prospector VII participants and sponsors wish to acknowledge the contributions of members of a prior meeting, the DOE/ARPA-sponsored Review Meeting on Fuel Cells for Direct Methanol Oxidation, Baltimore, MD, April 26-27, 1994. The attached summary of discussions from that two-day review proved useful in assisting the members of Prospector VII's Key Research Issues Working Group as they reviewed and prioritized issues in the DMFC category. ***That summary of discussions from the April 1994 meeting is hereby included as Appendix B to complete the Prospector VII Proceedings.***

For more information on this subject, please contact Los Alamos National Laboratory directly through one of the following individuals:

- a. Dr. Shimshon Gottesfeld, (505) 667-0853, FAX 665-2768
- b. Dr. Thomas Zawodzinski, (505) 667-0925, FAX 665-4292

Direct Methanol Fuel Cell Review Meeting April 1994

Executive Summary

Introduction

To establish the status and improve the understanding of critical issues relevant to research and development of direct methanol fuel cells (DMFCs), the Department of Energy (DOE), in cooperation with the Defense Department's Advanced Research Projects Agency (ARPA), convened a review meeting on April 26-27, 1994 in Baltimore, MD. The meeting was coordinated by Lawrence Berkeley Laboratory (LBL) and Los Alamos National Laboratory (LANL). The meeting brought together researchers and industrial fuel cell developers to provide a forum for scientific and technical exchange that promises to (i) assist the industrial developers in improving the performance of DMFCs, and (ii) identify areas of work in this field for research organizations that are currently active and where successes are needed to enhance the prospects for commercialization.

Fuel cells have attracted a great deal of attention for potential use in transportation applications, because of their good performance, high efficiency and low (or zero) emissions. A difficulty that has been recognized is in establishing the infrastructure needed to provide the fuel cell vehicles with alternative fuel. Methanol has attracted much interest as the fuel of choice because of its acceptable energy density and its compatibility with the existing liquid fuel infrastructure. The "conventional" approach for using methanol in fuel cells is to employ a reformer to produce hydrogen, which is consumed in the fuel cell. Since hydrogen made in this way contains sufficient carbon monoxide to poison the anode catalyst, the use of shift and preferential oxidation reactors to eliminate this poison is essential. These additional reactors lower the overall system efficiency and increase weight, volume and complexity. The desirability of direct oxidation of methanol in the fuel cell is clear, but much more research and development is needed before this can be considered to be practical. The results obtained to date have demonstrated the feasibility of direct electrochemical oxidation of methanol in laboratory-scale cells, but very significant improvements in the technology are still needed before a viable fuel cell technology of this kind is available for transportation applications. R&D programs sponsored by various government agencies have begun to address the critical technical issues facing DMFCs. There is still a great deal of improvement needed on several key issues before this important technology can be viable.

Status

The previous workshop was held on May 14-16, 1990 at Georgetown University, Washington University to discuss the R&D programs on direct oxidation of methanol and to formulate recommendations for advancing the technology. Since that meeting, there has been substantial progress. The performance of the anode for methanol oxidation has been improved, and there has been greater attention to DMFCs based on polymer electrolyte membranes.

Government-Sponsored Programs

The major support for DMFC technology comes from DOD/ARPA Army Research Office, and DOE/Office of Transportation Systems. Further substantial expansion by ARPA is planned in 1994 with a 4-year program to develop DMFCs for electric vehicle applications. The objective of this program is to demonstrate the DMFC technology for DOD vehicle requirements. The U.S. Army Research Office recently initiated a program to investigate anode electrocatalysts and also to develop membrane electrolytes with reduced

methanol permeation. The DOE program of exploratory research on DMFC is conducted mainly at LANL and at LBL. These programs are focused on studies related to electrocatalysis, improved electrolytes, and laboratory cell testing.

Summary and recommendations

A brief summary of the state of the art is given below for each of the main topics, followed by a summary of the information presented at the meeting for that topic. It should be noted that the principal problems for DMFCs, namely poor electrocatalysis and methanol crossover, impact on all of these topics. Recommendations were proposed by the participants and they are documented here.

An agenda for the Review Meeting and the List of Participants are provided in Appendices A and B, respectively. Information on specific individual presentations can be obtained by contacting the authors identified in the agenda (Appendix A).

Electrocatalysis

State of the Art. Electrocatalysis issues for DMFCs include the need for improvements in both methanol oxidation catalysts and methanol-tolerant oxygen reduction catalysts. The most promising anode catalyst of choice appears to be Pt-Ru, with variations in the nature of the particle supports and extent of dispersion. The state-of-the-art performance is summarized in Figure 1, which shows the intrinsic activity of various catalysts reported in the literature, normalized for temperature and surface area. Most of the data are for Pt alloys supported on carbon. The highest activity presented (by Kosek et al.) is for the "unsupported" catalyst consisting of Pt deposited on high-surface-area ruthenium oxide. The mechanism of methanol oxidation is understood to involve the initial dehydrogenation of methanol on the Pt surface to form adsorbed CO or other carbon-oxygen surface species. Additional components of the alloy catalyst serve as sources of active surface oxygen species which react with the CO to form CO₂. The details of these processes as well as optimized Pt/Ru ratios for catalysis and further characterization were presented by Markovic, Gasteiger, Ross, and Cairns for bulk alloy electrodes. This mechanism would predict first order in methanol concentration at low methanol concentrations, as the overall rate is controlled by methanol dehydrogenation at 'bare' surface sites, giving way to zero order at higher methanol concentrations as the bimolecular surface process $O_{ads} + CO_{ads} \rightarrow CO_2$ controls the overall rate.

In addition to the limitation of the anode catalyst, it is now clearly documented that MeOH permeation through the membrane in a DMFC with an ionomeric electrolyte brings about a significant loss of performance at Pt catalysts used in the DMFC air cathode. The reason for the loss is that, at the typical potential of the oxygen cathode (~0.8 V vs. RHE), MeOH is electro-oxidized effectively. The net cathode current is thus the current of oxygen electro-reduction minus the current of MeOH electrooxidation, where the latter component of the current leads to loss of cathode potential of the order of 100-150 mV at 100 mA/cm².

Summary of Discussion. The key issue is how to achieve stable, improved catalyst performance for methanol oxidation at the anode and oxygen reduction at the cathode. Practically, lower noble metal catalyst loading is required, particularly at the anode, as well as improved OCV's for methanol oxidation and minimizing the negative effects of methanol cross-over on cathode performance.

It was advocated by some participants that substantial additional effort be directed to fundamental studies of electrocatalysis, primarily on single crystal electrodes, to examine the intrinsic, non-poisoned activity of methanol electrooxidation. A typical turnover number for a heterogeneous catalyst is on the order of 10 reactions/Pt site-sec. For oxidation of methanol on Pt-Ru at 25°C, the turnover number is $\ll 1$, i.e. this is almost a 'dead' catalyst! However, extrapolation of transient currents measured at the clean surface of Pt electrodes to $t=0$ suggests substantially higher activities than are observed after poisoning. Also, different crystal faces of Pt have different intrinsic, clean surface activities, i.e. this is a "surface sensitive" reaction. The argument was made that the catalysis community could produce specifically oriented crystal surfaces even in dispersed catalysts. However, others disputed the relevance of this effort because concentration on measurements at short times for a pre-cleaned single crystal surface ignores the significance of the second, perhaps more relevant step in the mechanism at steady state, which involves the removal of adsorbed CO formed during the first step. Since the catalysts must last for thousands of hours in the DMFC, the consideration of the short term data for a specially pre-treated surface, even as a target, may be of little practical value. It was stressed that the relevant data, even in studies of single crystals and model interfaces, is the steady-state current density, since this most closely reflects the expected behavior of the real catalyst in the fuel cell. Furthermore, a predesigned surface structure is likely to change during long-term operation under fuel cell conditions.

Fundamental studies of the anode electrocatalyst using in-situ EXAFS and FTIR methods were described. Theoretical studies of relevant surface processes are also underway at CWRU.

An informal target for anode catalyst activity is provided in Figure 1 which suggests that 20 $\mu\text{A}/\text{cm}^2$ of real surface area is sufficient to achieve reasonable DMFC anode performance. However, performance increases beyond this level are very desirable to reduce the precious catalyst loadings that are now on the order of 4 $\text{mg}/\text{cm}^2/\text{electrode}$ in the DMFCs with the highest performance reported to date. The possibility of alternatives to the precious metal anode catalysts, such as metal macrocyclics, was raised. Unfortunately, reports to date indicate that the macrocyclics are inactive in methanol oxidation. While there has been some promise demonstrated for this class of catalysts in the cathode reaction, it was pointed out that methanol oxidation is an even more difficult task than oxygen reduction, and the rather poor results obtained in the latter case are not likely to be improved. One promising result was shown by Scherson and co-workers at CWRU. These workers have prepared carbon-supported Pt/polyoxometallate (e.g. $\text{H}_4\text{SiW}_{12}\text{O}_{40}$) methanol electrooxidation catalysts. At 148°C in phosphoric acid at moderate current densities, 50-150 mV improvements versus Pt/C were reported.

Clearly, the strong adsorption of the reaction intermediate, or of products of parallel processes, is the most important reason for the low rate of the methanol oxidation process and the effectiveness of an alternative catalyst depends upon its sensitivity to such surface poisoning effects. A better understanding and improvement of this vital step in the process is necessary to realize any improvement in alternative catalysts.

An entirely different approach in anode catalysis is to consider mediated electron transfer involving the homogeneous oxidation of methanol. DuPont reportedly pursued this approach but the efforts have not been published. Currently, Whitesides (Harvard Univ.) is investigating the mediated oxidation of methanol using Fe(III) in a separate reactor, then supplying the reduced species, Fe(II), to the fuel cell for electrooxidation to complete the loop. In work carried out thus far, a substantial voltage penalty is incurred and some adjustment in the redox potential of the mediating couple is needed to lower this

loss. However, it was warned that the problem is not only thermodynamic but also kinetic: any "fine tuning" of the redox potential to lower the voltage penalty will most probably also result in a slower homogenous oxidation reaction. A further criticism was that a precious metal catalyst was still required in the "off-line" homogeneous oxidation reactor, indicating that there is no apparent advantage gained at this point with this approach.

As far as the "conventional" Pt-Ru catalysts are concerned, the ARPA project uses a high precious metal loading of about 4 mg/cm². This is not necessarily the lower limit of the practical loading for advanced anode catalysts. Further work is needed to replicate the successes of the hydrogen/air fuel-cell community, that is to lower the loadings substantially without penalizing the performance. A viable fuel cell for transportation applications will not be able to tolerate the high costs of such loadings of Pt-Ru.

The other area to concentrate on in electrocatalysis is methanol-tolerant cathode catalysts. Unless a methanol impermeable barrier is present in the fuel cell to limit cross-over of the fuel from the anode to the cathode, there will be a penalty because of methanol oxidation at the cathode catalyst. Therefore, catalysts which are methanol tolerant, i.e. do not suffer a performance limitation in the presence of methanol, should be an additional target of research. A possible class of effective cathode catalysts in this regard are high-surface area, electronically conducting metal oxides. CWRU observes that carbon-supported high-surface area RuO₂ is a poor methanol oxidation catalyst, and, while not the equal of Pt based catalysts, is relatively active as an oxygen reduction catalyst. A substantial gain in DMFC performance can be obtained with Pt cathode catalysts by appropriate tailoring of the electrode structure. Apparently, the Pt based cathode used in the ARPA project (Kosek et al.) is somehow structured such that it is relatively tolerant to methanol.

Polymer Electrolyte and Alternative Electrolytes

State of the Art. A key target in the selection of the cell electrolyte is the minimization of methanol cross-over. A number of electrolytes were discussed. Recently, the most commonly used electrolyte is Nafion 117, a poly(perfluorosulfonic acid) (PFSA) material. Use of this, as opposed to other PFSA materials which have better performance in hydrogen-based fuel cells, is driven by the more widespread availability of Nafion 117 and its relatively greater thickness. The methanol flux through the membrane is, to first approximation, proportional to the inverse of the thickness of the membrane. Another electrolyte which has been investigated is the aqueous buffered carbonate electrolyte. One of the primary advantages is the low solubility of methanol in the high-temperature carbonate electrolyte. Various forms of composite membranes could be beneficial to lower the degree of methanol crossover in DMFCs based on membrane electrolytes. One such class of electrolytes is the composite membranes prepared from tin mordenites (a zeolitic structure) supported in poly(acrylic acid) membranes. While the conductivity of the composite material is not particularly high, a decrease in the methanol cross-over rate by a factor of two relative to Nafion 117 was reported.

It is important to keep in mind that the central membrane requirements of hydrogen fueled fuel cells, i.e. high chemical stability and high protonic conductivity have to be met together with the special requirements of low methanol crossover and higher temperature operation, by any electrolyte for DMFCs.

Summary of Discussion. Approaches to limiting the methanol cross-over can involve, in principle, some combination of lowering its solubility in the electrolyte and inhibiting its

mobility through the electrolyte. An important point is that all such attempts involve tradeoffs, i.e., to accept somewhat lower electrolyte conductivity (especially in light of the lower current densities in DMFCs) in exchange for improved barrier properties toward methanol. Lower solubility of methanol can be achieved by either physical modification of the membrane electrolyte (e.g. cross-linking) or by using the tendency of the membranes to lose imbibed liquid at elevated temperatures. Unfortunately, for materials such as PFSAs, which require water to maintain adequate ionic conductivity, higher temperatures can be deleterious since water as well as methanol is lost from the membrane. Furthermore, overall material stability is more difficult to maintain at high temperatures.

Some attempts are underway to prepare suitable materials which maintain satisfactory conductivity above 150°C, with an especially strong effort taking place in the context of the ARPA-sponsored program centered at CWRU. One class of materials under development have typically involved using phosphoric or sulfuric acids to provide conduction in the membrane at elevated temperatures. The matrix materials used are Nafion and poly(benzimidazole). Additionally, new materials such as perfluorinated sulfonimides, which are strong acids, are being synthesized. The latter materials may be more thermally stable than sulfonic acids. A fundamental problem with ionomeric materials is separation of microphases (i.e. ionic pores and backbone regions). Not only can the hydrophobic backbone regions potentially serve as methanol conduits, but the regions with ionic conductivity also inherently attract methanol. The development of "single phase" structures is not without its difficulties. Such structures either provide poor conductivity or are not sufficiently stable (e.g. polybenzimidazole with phosphoric acid). All in all, preparing single phase flexible methanol rejecting polymers with adequate conductivity is a daunting task. While new materials based on sulfonimides are being investigated, the properties of membranes derived from these materials must be critically compared with existing membranes. There is little reason to believe *a priori* that sulfonimides promise particularly advantageous properties *vis a vis* methanol cross-over.

The development of an anion-conducting, CO₂-rejecting membrane which could operate with neutral or basic internal pH would have significant advantages as a membrane for direct methanol (or even hydrogen) fuel cell applications. Cathode electrocatalysis would be improved. However, existing membranes have poor stability at reasonable temperatures under basic conditions. Several aqueous systems which have some of the properties of anionic membrane systems were described at this meeting. For example, an aqueous carbonate system with a Pt-Ru catalyst yields a very substantial gain in performance compared to a cell with a phosphoric acid electrolyte. This anode provides 100 mA/cm² at 300 mV overpotential at atmospheric pressure and 120°C utilizing a methanol vapor feed. A significant limitation of the electrolyte is that the solubility of oxygen is not very high. On the other hand, the methanol solubility is also quite low, thereby reducing methanol crossover.

Inorganic membranes or organic/inorganic composites could offer some advantages. The protonic conductivity of most inorganic membranes in the moderate temperature range (100°C to 200°C) is typically low. Hydrated complex salts, which can exhibit fairly good conductivity, are usually metastable. One system which has been shown to have fairly good conductivity with some methanol rejection is a composite membrane prepared from tin mordenite (an aluminosilicate) particles in an acrylic acid/methacrylate co-polymer matrix. It is unclear what the eventual stability of such a membrane will be in the fuel cell environment. It is quite likely that the decreased methanol permeation rate is simply an effect of increased tortuosity because of the zeolite particles introduced into the matrix. Another inorganic component, incorporated into the membrane to lower methanol permeation, was demonstrated at LANL to provide approximately 50-

mV improvement in performance of a Nafion 117-based DMFC over most of the polarization curve.

Cell Issues

State of the Art. The best state-of-the-art DMFC performance currently reported by JPL is a liquid-feed (2 M methanol in water) cell which delivers 230 mA/cm² at 0.5 V with an air cathode. This cell utilizes a Nafion 117 membrane, a Pt-Ru unsupported anode catalyst (4 mg Pt-Ru/cm²) and a specially designed cathode. Fuel cells based on aqueous carbonate electrolytes operating at higher temperatures have delivered 640-670 mV at 100 mA/cm² in a rather complex pressurized system. The cells operate at 165°C and the system utilizes CO₂ recycling from the anode effluent to the cathode feed to minimize pH variations in the cell. A total precious metal loading of 0.5-1 mg/cm² was utilized. The performance of anode catalysts with various Pt/Ru ratios was reported for this aqueous carbonate cell. The methanol cross-over was low. There was some concern with metal corrosion in the cell hardware as Ni components are apparently susceptible to the alkaline electrolyte at the elevated temperature.

Summary of Discussion. Questions were raised concerning the cost of DMFC components such as backing materials and the manufacturability of these cells. A component manufacturer present in the meeting indicated that the automation and scale-up of some of the manufacturing processes could significantly lower cost.

The issue of methanol crossover and how to deal with it is a critical issue at the cell level. One solution to this problem at the cell level is the development of methanol-tolerant cathode catalysts. It was reported that FeTMPP supported on carbon exhibited reasonable performance for oxygen reduction based on SNL tests in alkaline electrolytes. However, previous LANL results had suggested that pyrolyzed macrocycles were rather poor oxygen catalysts in acidic PEFCs. The additional losses in the oxygen reduction reaction encountered with such non-precious metal catalysts could negate any expected improvement to be gained by the increased tolerance of the catalyst to methanol.

The issue of alternate fuels was broached. Substantial work has also been carried out by JPL and collaborators on oxygenated fuels. However, the cost of these fuels is likely to be high. Methane and hydrocarbons in general are poor candidates for electrochemical oxidation at modest temperatures. Ammonia is not considered viable from a toxicity standpoint. On the other hand, it was also pointed out that problems related to methanol toxicity are probably overrated.

Extensive modeling may allow assessment of the variety of methanol cross-over effects at the cell level. The relevant transport parameters will need to be experimentally derived. With an effective cell model, the efficacy of various options and modifications can be evaluated, for example, the relative merits of vapor and liquid feed, the effects of modifying the electrode structure and its optimization, improved understanding of the effects of pressurization on electrode performance (i.e., is the improvement in cathode performance from pressurization an effect of methanol removal from the cathode backing or merely an increase in oxygen partial pressure?).

Stack/System Issues

State of the Art. IFC has investigated relatively small direct methanol fuel cell stacks under the ARPA program. Both air and oxygen cathode feeds have been implemented. Performance nearly equivalent to that of a single cell is attained upon scale-up to a stack.

Methanol crossover is measured by analyzing for CO₂ present in the cathode effluent. A current-density equivalent to approximately 50 mA/cm² of methanol is lost to crossover when the fuel cell is operated at a current density of 250 mA/cm² with a lean feed. The stack is cooled by circulation of the liquid feed.

Summary of Discussion. Several of the issues addressed here contained elements of the earlier sections, such as the system impact of crossover problems and the resultant efficiency penalty. It was questioned whether the thermal load could be adequately addressed by the cooling of the recirculated anode feed. Pressurization appears to be necessary to attain higher performance and minimize the effects of crossover. Can satisfactory performance be attained with the use of air and/or unpressurized reactants? Is operating the cells at higher temperatures a viable option in stacks? Answering these questions calls for a definition of system requirements in conjunction with modeling of the system in order to examine the relative efficacy of the various approaches.

If the liquid feed is recirculated through the stack, a certain amount of methanol will be lost in the CO₂ product stream because of its high volatility. If the elimination of emissions (e.g. the residual methanol or any other possible by-products of the anode process) becomes an important issue, the recovery or destruction of the residual vapor in the anode effluent may be required. One possible solution is to mix the effluent stream with air and run it over an oxidation catalyst.

A possible concern in liquid-feed fuel-cell stacks is the shunt losses arising from ionic conduction through the feed, although they would certainly be less than that experienced in battery systems (such as Zn/air) that use highly conductive anolytes.

Recommendations

A set of recommendations was proposed by the attendees at the Review Meeting on April 26-27, 1994. These recommendations are documented and address several topical areas, including electrocatalysis, polymer electrolytes and alternative electrolytes, cell issues, and stack/system issues.

ELECTROCATALYSIS

The catalyst loadings presently used to obtain reasonable performance in DMFCs are excessive from a systems cost perspective. Therefore, anode catalyst activity has to be increased by a factor of 10 to be able to lower precious metal loadings by an order of magnitude.

(a) Further improvements in the performance of the best anode catalysts identified to date, i.e., Pt-Ru, can be achieved by optimization of the catalyst structure, particularly by increase of the surface area in stable structures, and optimization of the structure of the catalyst layer. The latter is of great importance, as combined requirements of protonic conductivity, electronic conductivity, liquid access (MeOH) and gas permeability (CO₂ product) need to be met.

(b) Fundamental studies of the intrinsic electrocatalytic properties of model interfacial systems, e.g., single crystal electrode or bulk alloy electrodes, can advance the understanding of the anodic oxidation of MeOH. It is recommended that steady-state currents be reported for such model systems during electrooxidation of MeOH to improve the relevance to "real life" DMFC experiments.

(c) Pt/Ru-Oxide ("unsupported Pt-Ru") catalysts have been demonstrated recently as highly effective in DMFCs. Fundamental studies of MeOH electrooxidation at relevant Pt-RuO_x model systems are recommended.

(d) It is highly desirable to know the surface composition and structure of dispersed alloy catalysts, particularly Pt-Ru, in DMFC anodes. Information at this time is confined to the overall bulk composition, and information on the surface composition of the catalysts in the DMFC anode is lacking. It is recommended to apply techniques such as EXAFS to obtain information on catalyst surface composition/structure in operating DMFCs.

(e) Effective, MeOH-tolerant cathode catalysts for DMFCs are highly desirable. The prerequisite should be an activity corresponding to a cathode potential lower than that of Pt by not more than 50 mV @ 100 mA/cm² of ORR current density in the absence of any interference. Otherwise, the MeOH tolerance will not compensate for the lower intrinsic performance.

(f) The deleterious effects of MeOH on the ORR at the Pt cathode catalyst can, in principle, be prevented by smart designs of the DMFC cathode. Such efforts are recommended.

POLYMER ELECTROLYTE AND ALTERNATIVE ELECTROLYTES

The use of membrane electrolytes in DMFCs has been the most significant new development in this technology. However, MeOH crossover through the electrolyte is a major problem. For many applications, a fuel efficiency of only 50% at a current density of 100-200 mA/cm² is unacceptable, particularly for transportation applications.

(a) Modify existing polymeric membranes by introducing an interpenetrating network or other composite structure while demonstrating long-term stability of the structure under operating DMFC conditions.

(b) Develop new membranes that would combine satisfactory conductivity with low MeOH permeability.

(c) Demonstrate long-term stability and good cell performance for phosphoric acid-soaked membranes, as well as for other similar systems, operating at higher DMFC temperatures.

DMFC CELLS

The report of a performance of ca. 300 mA/cm² @ 0.5 V for MeOH/O₂ cells employing membrane electrolytes is quite exciting. However, this is still quite far from the requirements for a viable cell performance for transportation applications, and the encouraging results have not yet been demonstrated on a time scale longer than several days.

(a) Demonstrate stable cell performance with high anode loadings on a time scale of several weeks, then several months.

(b) Lower anode catalyst loadings and demonstrate stable performance over time periods extending to months.

(c) Compare performance in cells using liquid and vapor feeds of methanol.

(d) Carry out tests over the widest temperature range possible with ordinary ionomeric membranes.

(e) Develop a comprehensive DMFC model to predict the effects of MeOH crossover on cell performance and cell efficiency, to compare liquid vs. vapor anode feed performance, etc.

DMFC STACKS

A DMFC stack that is fed directly by liquid MeOH is very attractive for transportation applications as well as for other applications which may utilize the complicated system of a methanol reformer coupled to a H₂/air PEFC. However, the cost per kW, the specific power and the fuel efficiency of a DMFC stack are still far from satisfactory for transportation applications. Therefore, it may be debatable if it is appropriate to develop DMFC stacks before an advanced technology is available which overcomes some of the above challenges. The general feeling at the meeting was that such efforts are a worthwhile contribution to the general development of fuel cell manufacturing capabilities in the United States.

(a) Develop stack and system concepts based on DMFCs with liquid or vapor feeds. Strive to lower cost, increase power density, and demonstrate long-term performance to levels where comparison with the MeOH reformer + H₂/air PEFC stack will be more clearly in favor of the DMFC.

(b) Develop a reliable computer model of DMFC stack performance.

(c) Operate DMFC stacks to probe emissions to determine if they qualify for a ZEV or, at least, for a ULEV.

Institutional Considerations

The introduction of an alternative fuel to the transportation sector presents a number of issues that must be considered, such as:

- safety considerations
- storage characteristics
- availability
- new infrastructure requirements
- environmental considerations
- compatibility with fuel cell systems

These issues have, to some extent, been considered in the existing DOE Program on Fuel Cells for Transportation, but there is still a need to completely resolve some of them.

Methanol was selected as the fuel of choice for direct-hydrocarbon fuel cells in transportation applications because it can be derived from non-petroleum sources, is easy to transport and store, and offers the best prospects for obtaining reasonable efficiency from the direct electrochemical oxidation of a liquid fuel. Furthermore, the existing refueling infrastructure could be readily adapted to handle methanol for use in either indirect (reformed) or direct methanol fuel cells. Indeed, a number of existing gas stations have been modified to offer M85 (85% methanol, 15% gasoline). Methanol has toxic properties which must be addressed when it is used as a transportation fuel. The relatively high volatility of methanol compounds the toxicity issue and could conceivably exacerbate the

occurrence of potentially explosive situations in which vapor accumulates in an unvented area. The use of methanol is of concern from an environmental standpoint because it is infinitely miscible with water and spillage could contaminate aquifers and other water storage reservoirs. These issues have been addressed to a large extent elsewhere and it is generally concluded that safety in storing and handling methanol can be resolved with the use of appropriate procedures. Many of the problems are similar to those of other transportation fuels, and they have proven amenable to practical solutions.

Institutional issues specifically related to the safety and handling of methanol for both DMFCs and fuel cells that utilize hydrogen from steam-reformed methanol should be similar. In these systems, substantial escape of methanol may occur, especially in the case of the DMFC where appreciable evaporation from the edges of the membranes may occur in current stack designs, and cross-over may result in methanol in the cathode effluent stream. Perhaps of greater concern is the possibility that undesirable by-products, such as formaldehyde, dimethyl ether, etc., might be formed in the DMFC or in the fuel processing system of the reformed-methanol fuel cell system. In the case of the DMFC, these difficulties may be compounded by higher temperature operation.

The most significant research needs related to institutional considerations are:

- (a) Investigate the evaporative losses of methanol in DMFCs.
- (b) Identify undesirable by-products that may form in the fuel cell and which could be vented from the system.

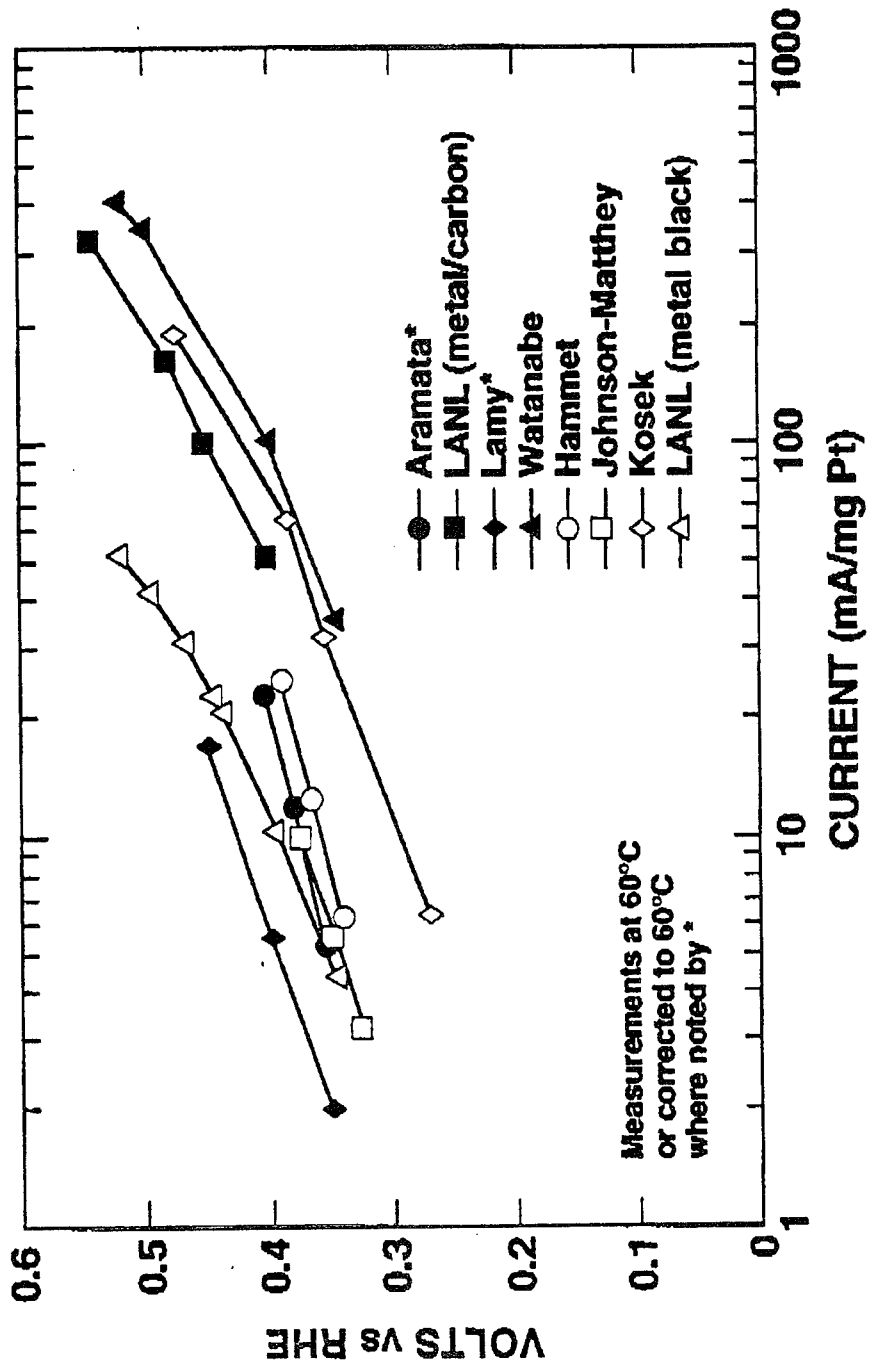


Figure 1.

Agenda

Fuel Cells for Direct Methanol Oxidation Review Meeting

April 26-27, 1994

Sheraton International Hotel
Baltimore/Washington Airport

April 26, 1994

Technical Presentations

- 8:30-9:00 A. Landgrebe (DOE) - Introductory remarks
9:00-9:30 N. Markovic (LBL) - Electrocatalysis of MeOH Oxidation on Pt-Ru Alloys
9:30-10:00 W. O'Grady (NRL) - Experimental Studies of Methanol Oxidation
10:00-10:15 Break
10:15-10:45 J. McBreen (BNL) - Platinum Alloy and Pt/UPD Metal Electrocatalysts:
In Situ XAS Studies
10:45-11:15 D. Scherson (CWRU) - New Approaches to the Design and Optimization
of Pt-Ru Electrocatalyst for Methanol Oxidation
11:15-11:45 D. DesMarteau (Clemson Univ.) - Novel Electrolytes for Direct Methanol
Fuel Cell
11:45-1:15 Lunch
1:15-1:45 M. Litt (CWRU) - Acid Doped Polybenzimidazole Electrolytes for the
DMFC
1:45-2:15 S. Gottesfeld (LANL) - R&D on PEM Fuel Cells for Methanol Oxidation
2:15-2:45 R. Surampudi (JPL) - Direct Methanol Fuel Cell Technology
2:45-3:00 Break
3:00-3:30 S. Srinivasan (TAMU) - Methanol Oxidation in PEM Fuel Cells
3:30-4:00 J. Kosek (Giner, Inc.) - Direct Methanol PEM Fuel Cell Tests
4:00-4:30 D. Maricle/D. Wheeler (IFC) - Development of Direct Methanol Fuel Cell
Stack
4:30-5:00 E. Smutkin (IIT)/D. Zurawski (ANL) - PEM DMFC Program

April 27, 1994

Formulating Recommendations

- 8:30-10:00 S. Gottesfeld - Discussion Leader: Formulating R&D Recommendations
10:00-10:15 Break
10:15-12:00 S. Gottesfeld - Discussion Leader: Documenting
Recommendations/Wrapup
12:00 Adjourn

**Attendees List - DMFC Review Meeting
Baltimore, MD
April 26-27, 1994**

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