

PROCEEDINGS OF THE WORKSHOP ON

**DIRECT METHANOL-AIR
FUEL CELLS**

Edited by

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ENERGY TECHNOLOGY AND PHYSICAL ELECTROCHEMICAL DIVISIONS

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**DIRECT METHANOL-AIR FUEL CELL:
AN OVERVIEW OF THE WORKSHOP**

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1.0 INTRODUCTION:

Fuel cells are efficient energy conversion devices that convert chemical energy into electricity. Hydrogen is the commonly used fuel in these devices and oxygen from air is the oxidant. For space applications, the hydrogen fuel is stored on board or produced via the electrolysis of water. Currently electrolytic hydrogen is not cost effective for terrestrial application. Thus, the hydrogen is obtained by "reforming" a variety of fossil fuels such as natural gas. In a reformer, the fossil fuel reacts with steam at elevated temperatures in the presence of selected catalysts to produce hydrogen, carbon dioxide and small amounts of carbon monoxide. Extreme care must be taken to ensure that the amount of carbon monoxide produced is kept to an absolute minimum as the CO has a strong poisoning effect on the anode catalyst (platinum) in both phosphoric acid (PAFC) and proton exchange membrane fuel cells (PEMFC).

The molten carbonate (MCFC) and solid oxide fuel cells (SOFC) do not suffer from this limitation, since their high operating temperatures negate the poisoning effect of CO. In fact, these systems in principle do not need the reformer as the hydrocarbon fuel can be internally reformed within the fuel cell at the operating temperatures of these systems. However, the elevated temperatures limit the applications of these systems primarily to utility applications. They are not suitable for transportation and a variety of stand-alone applications.

A fuel cell operating at between 100 deg.C. to 200 deg.C and using a hydrocarbon fuel directly provides two significant benefits. The first is that it eliminates the need for a reformer, thus reducing the complexity and size of the system as well as the cost. The size reduction is particularly important in applications such as transportation where packaging the propulsion system in the close confines of a vehicle is a paramount concern. In addition, some estimates indicate that the reformer and its auxiliary support instrumentations can cost as much a third of the total system. A cost reduction of this magnitude is very important in both stand-

alone and transportation applications and can make the difference in the commercial viability of a fuel cell. The second advantage arises from a well known limitation of most reformers. During steady state operation they demonstrate a high degree of efficiency. However, under transient loads, their conversion efficiency decreases substantially resulting in the enhanced production of unwanted byproducts such as carbon monoxide. By eliminating the reformer one can design systems which are very responsive to transient loads experienced in vehicular applications.

Despite these advantages developing such a fuel cell remains a daunting task. Catalysts that either promote internal reforming of hydrocarbons at these lower temperatures or are capable of direct electrochemical oxidation of such hydrocarbons are non-existent. With an alternative fuel like methanol, the situation is still challenging but appears to be more tractable than with hydrocarbons. Recent research indicates that promising catalysts for direct electrochemical oxidation of methanol may indeed be feasible. Thus, programs to develop a direct methanol-air fuel cell are in progress in Europe and Japan. The recent enhanced interest in the U.S. to promote the use of methanol for transportation applications warrants a thorough examination of the potentialities of a direct methanol-air fuel cell for such applications.

A three day workshop (May 14 - 16, 1990) on Direct Methanol-Air Fuel Cells was held at the Leavey Center of Georgetown University. The objective of the meeting were

1. to review the current status of research,
2. and based on the review, to identify areas that need further research and development.

To accomplish the dual objectives the agenda included formal presentations by experts on current research on methanol-air fuel cells and its components. The formal presentations were followed by group discussions on research needs. On the final day of the workshop, the group moderators presented the recommendations of the groups to all the participants of the workshop. The agenda of the workshop is attached (Table I).

The workshop focussed on several important areas including:

1. research on the kinetics and mechanism of methanol electro-oxidation,
2. the use of advanced surface characterization techniques to identify reaction intermediates,
3. appropriate electrolytes for a direct methanol-air fuel cell.

Issues relating to cell design and testing were not addressed in much detail since it was clear from the workshop that the technology is in its infancy and that research on cell design can only begin following the identification of promising catalysts for the anodic oxidation of methanol and the selection of a suitable electrolyte. However, the workshop did recognize the need for an overall systems perspective even at this early stage so that the research on the various components can proceed with guidance from the systems analysis.

The meeting was well attended with some 50 participants from academia, federal laboratories and industry. A strong contingent of foreign visitors was present at the meeting, five from Japan and nine representing various European countries. A complete list of attendees is provided in Appendix I. The substantial participation from Japan and Europe reflects the fact that direct methanol-air fuel cell is considered to be an attractive technology by our main trading partners. It is also important to note that the Japanese and European efforts are more comprehensive than the current DOE effort in direct methanol-air fuel cell technology.

This paper summarizes the status of research in direct methanol-air fuel cells and proposes a research agenda in this important technology based on the consensus that evolved in this workshop. It is important to note that the proposed program is considered to be complementary to the existing DOE program to develop PEM fuel cells operating on reformed methanol for vehicular applications. The direct methanol-air fuel cell, if successful, will evolve as a second generation technology to the PEMFC.

2.0 STATUS:

The direct methanol-air fuel cell is in a much earlier state of development than other fuel cell systems such as PAFC and PEMFC. The most significant program in this technology is supported by the Commission of the European Communities (CEC) and involves a coordinated program involving several European governments and industry. The program builds on the foundations established in earlier programs at EXXON and Shell in the early 1970's. The participants in this program are listed in Table II.

The program emphasis has been on developing an understanding of the mechanism of methanol electrooxidation and on using this knowledge to identify high-performance anode catalysts with long life for use in the fuel cell. Progress in this endeavor was summarized at the workshop by Dr. Cameron of Johnson Matthey. The Platinum/Ruthenium catalyst has allowed this consortium to improve the anode catalyst activity from 5-6mA/mg of Pt to approximately 50mA/mg. This is a significant accomplishment. However, this in itself is not adequate for the direct methanol-air fuel cell. A further improvement of two to five fold is required. The lifetimes of these catalysts have also not been evaluated adequately. Overall

this program has demonstrated a power density of 50mW/sq.cm. at a catalyst loading of less than 1mg/sq.cm. at operating temperatures of around 80 deg. C.

Table II CEC Fuel Cell Program

Participant	Research Area
Johnson Matthey (UK)	Catalysts and Cell Evaluation
Univ. of Oxford (UK)	Mechanistic Studies of Anode Reactions
Univ. of Southampton (UK)	UHV studies of Catalysts
CNRS (France)	Mechanistic Studies of Anode Reactions
Univ. of Poitiers (France)	Chromatographic and IR Reflectance Studies of Catalysts
Univ. of Bonn (Germany)	Co-catalysts for methanol Oxidation.
Siemens (Germany)	Catalysts and Cell Evaluation
Univ. of Cork (Ireland)	Air Electrodes

The Japanese effort in this system involves Hitachi which is supported in terms of fundamental research by Dr. Watanabe at Yamanashi University and the Equos Research Inc. The concept proposed in this program utilizes the PEM concept where catalysts are impregnated on an ion exchange membrane with solubilized Nafion. The catalysts used are Platinum/Ruthenium and Platinum/Tin. Steady state operation in excess of 150 hours is claimed at current densities in the range of 100 mA/sq.cm. This performance is still quite inferior to PEMFC's operating on reformates; however, it is very promising compared to what has been achieved with this system in the past. Like the European effort this program is also devoting significant resources to understand the mechanism of methanol oxidation and to identify better performing and cost effective catalysts for the reaction.

Unlike the European and Japanese efforts the U.S. at present does not have a focussed program in direct methanol-air fuel cells. The Office of Naval Research (ONR) has been supporting academic researchers in the study of the kinetics of methanol oxidation reactions. A variety of advanced surface sensitive techniques is being used to understand the atomic level details of methanol adsorption and its subsequent reactions on catalyst surfaces. Progress is being made in identifying the reaction intermediates,

as well as in developing an understanding of how to control the formation of unwanted intermediates. Recently, ONR and DARPA joined forces to initiate a program on direct methanol-air fuel cells under the auspices of the Jet Propulsion Laboratory (JPL).

The only DOE supported program in this area is the work done at Giner Inc. under a Phase I contract of the SBIR program. Unlike the European and Japanese approaches, Giner Inc. employed a buffered carbonate electrolyte. The fuel cell was operated at elevated pressure and temperatures up to 180 deg.C. Performance of 550 to 600mV at 150 mA/sq.cm. was obtained at 120psig and a temperature range of 165 to 180 deg. C. Decreasing the pressure to 80psig did not adversely affect performance. The catalyst employed was platinum at loadings of 0.5mg/sq.cm. for both the anode and cathode. An asbestos matrix and a cesium carbonate electrolyte were utilized. Periodic carbon dioxide additions to the electrolyte were needed to prevent any significant pH gradients within the cell.

The status of the direct methanol-air fuel cell research and development effort clearly indicate that despite the potential promise of this technology many significant technical hurdles still must be overcome. It also shows that the present efforts are systematically addressing major issues relating to catalysts and electrolytes. The progress to date suggests that continued research could lead to the required breakthroughs which would make this system commercially viable.

3.0 RECOMMENDATIONS OF THE WORKSHOP:

In order to develop a consensus research agenda for a direct methanol-air fuel cell, the participants of the workshop were divided into four working groups in the following areas:

1. Kinetics and Mechanism of Methanol Oxidation.
2. Surface Sensitive Techniques for Methanol Electrocatalysts.
3. Electrolytes for Methanol-Air Fuel Cells.
4. Systems Considerations for Methanol-Air Fuel Cells.

Each group had a moderator who facilitated the discussion and presented the recommendations of the group to all the workshop participants on Wednesday May 16, 1990. The groups addressing issues relating to Kinetics and mechanism (Group 1) and Surface Sensitive Techniques (group 2) agreed to meet in a joint session since many of the discussion topics in the two groups overlapped. However, the recommendations were presented separately by the two moderators.

The following sections summarize the recommendations of each of the groups:

3.1 Kinetics and Mechanism of Methanol Electro-Oxidation:

The general consensus was that progress in understanding the mechanism of methanol electro-oxidation has been good. A combination of kinetic and spectroscopic techniques has allowed researchers to identify a majority of the intermediates involved in this reaction. However, the group concluded that much remains to be done. It is clear that a practical catalyst for methanol electro-oxidation is still not available and that the search for such a catalyst must include: 1) the development of a detailed understanding of the kinetics and mechanism of the reaction under conditions relevant to direct methanol-air fuel cell operation and 2) the evaluation of new materials as potential catalysts for the methanol oxidation reaction. The specific recommendations in each of these categories are summarized below:

1.0 Kinetic and Mechanistic Studies:

1. Determination of conventional rate constants over a wide range of temperatures and pressures for a variety of catalytic systems.
2. Spectroscopic studies to identify reaction intermediates at temperatures close to fuel cell operating conditions.
3. Increased emphasis on kinetic measurements under steady state conditions on high area catalysts.
4. Integration of kinetic studies with electrolyte development for the direct methanol-air fuel cell.
5. Understanding the mechanism of CO formation and its dependence on surface structure and composition. The focus will be on understanding the dynamics of catalyst poisoning.
6. Understanding water and OH adsorption in model systems. Developing in-situ spectroscopic techniques for studying OH adsorption.

2.0 New Materials as Electrocatalysts for Methanol Electro-Oxidation:

1. Develop novel methods for preparing supported high-area multi-component catalyst materials.
2. Explore electronically conducting oxides (bronzes and perovskites) and carbides as substrates.
3. Evaluate oxides and mixed metal/oxide catalysts.

4. Evaluate ordered alloys such as Pt₃Ti.
5. Explore analogues of homogeneous catalysts for methanol oxidation.
6. Coordinate catalyst development with electrolyte development.

3.2 Surface Sensitive Techniques for Methanol Electrocatalysts:

A variety of surface sensitive techniques is now being used to examine electrochemical reactions. Papers were presented at the workshop where molecular level understanding of the mechanism of methanol oxidation was being developed by the use of both ex-situ and in-situ surface sensitive techniques. The results clearly show that it is useful to use a variety of techniques to obtain the different types of information required to understand the complex phenomena of methanol electro-oxidation. The working group focussed on the molecular level understanding that is required and did not attempt to identify specific techniques to obtain that information.

The group considered three distinct issues where surface sensitive techniques can play a major role. These are:

1. Mechanism of the Methanol Oxidation Reaction: There is a strong need to identify the nature, amount and most importantly, the time variation of the formation and decay of reactive intermediates, including poisons and the desired products of the reaction. A variety of spectroscopic techniques is being used for this purpose and the group recommended that such activities should continue.
2. Catalyst Characterization: The group recommended that emphasis be placed on characterization of single crystal, UPD and dispersed catalysts. These studies will allow us to understand the surface characteristics that are well suited for methanol electro-oxidation. Specific studies should include:
 1. In-situ characterization of single crystals particularly the high index planes and surface restructuring.
 2. UPD layers, particularly the interaction between the UPD metal, oxygen and the substrate. These studies should be conducted under full and partial coverages.
 3. OH adsorption.
 4. The adsorption of C₁ species and their diffusion on

the surface.

5. The morphology of dispersed catalysts, including catalyst-substrate interactions and alloy catalysts as well as UPD species. In addition, with dispersed catalysts, it is necessary to investigate electronic structure effects associated with small metallic clusters.
6. The careful study of well defined model systems using UHV systems. The effect of variation of the work function and the simulation of electrochemical conditions in these studies will provide fundamental insights into the mechanism. In addition, the mechanics and energetics of H_2O and OH adsorption from non-aqueous systems are also desirable.

3.3 Electrolytes for Methanol-Air Fuel Cells:

The group analyzed the merits/demerits of various electrolytes for a direct methanol-air fuel cell. The potential electrolytes considered in this analysis were:

1. Inorganic Acids such as H_2SO_4 , $HClO_4$, and H_3PO_4 .
2. Superacids such as TFMSA.
3. Buffers such as CO_3^{2-}/HCO_3^- .
4. Solid Electrolytes, including proton and oxide ion conductors.
5. Polymer Electrolytes such as perfluorinated sulfonic acids.
6. Alkaline Electrolytes such as $NaOH$.
7. Molten Carbonate Electrolyte.

The properties of these electrolytes were then compared to the requirements of the methanol-air fuel cell. This analysis is shown in a tabular form (Table I). A rating of 1 indicates that the electrolyte is well suited in terms of the fuel cell requirement. This matrix analysis allowed the group to prioritize the electrolyte requirements of the direct methanol-air fuel cell. The group recommended that the buffered systems, polymer electrolytes and the solid electrolytes are well suited for this application. In contrast, the superacids, molten carbonate and alkaline electrolytes are not particularly advantageous for this type of fuel cell. However, several participants felt that it was premature to reject the superacids, particularly in view of the recent

progress made with these electrolytes.

The group also recommended that research on the electrolytes needs to focus on the following issues:

1. Reactant/Product Management, including methanol transport/solubility and water management.
2. Stability of the electrolyte. For example, cross linking in the proton exchange membrane and developing similar membranes for elevated temperature operation.
3. Compatibility with anode kinetics and catalysis, including research on additives to improve anode kinetics.

3.4 Systems Consideration for Methanol-Air Fuel Cell:

The primary advantage of the direct methanol-air fuel cell is that it eliminates the need for a reformer. Therefore it potentially could be a simpler system with lower cost and footprint. It could also be better suited to respond to transient loads. These benefits are valuable for a variety of applications requiring 500W to 50KW units. These applications include both transportation and some stationary applications. However, even for these applications this group felt that major breakthroughs in catalysts and higher temperature electrolytes are needed if the direct methanol-air fuel cell is to compete with fuel cells operating on reformates.

The group also felt that the direct methanol-air fuel cell must demonstrate the well known common benefits of all fuel cells such as high efficiency, low pollution and unlimited operating time. There was some concern that the direct methanol-air fuel cell may have to sacrifice some of its simplicity in order to incorporate ancillary equipments such as burners for the removal of residual methanol and/or formaldehyde from the fuel cell stack.

The group strongly recommended that an initial systems analysis be performed to identify the trade-offs that are necessary for this system to be competitive. Sensitivities to catalyst improvements, higher operating temperatures and the impact of pressurized operation should be analyzed. This systems trade-off analysis could then form the basis of setting developmental goals for the program. Research progress should then be measured against these goals.

The group recommended a "stepped" ten year plan to develop the direct methanol-air fuel cell program. The plan should call for a small initial investment in research to address some of the issues

identified in this workshop. Increase in funding and in the scope of the program should be linked to accomplishments in the ongoing program. The investment of R&D dollars for this program should be balanced against the potential benefits and the technical hurdles identified in the trade-off analysis.

4.0 CONCLUSIONS:

The direct methanol-air fuel cell eliminates the need for a reformer, thus reducing cost and enabling the complete system to be compact. In certain applications such as transportation these are major benefits. However, the performance of this system at present is much inferior to that of either the PAFC or PEMFC. Unless the performance is significantly improved the need for additional cell stack with the direct methanol-air fuel cell to deliver the required power may negate the advantages of eliminating the reformer. It is not clear at this time what performance targets are required for this system to show substantial advantages over technologies being developed today. There is a great need to initiate a detailed systems analysis study to quantify these needs and establish the required performance goals of the direct methanol-air fuel cell. These goals will then serve as benchmarks to direct the R&D program and measure its success.

Parallel to the systems analysis effort, it would be prudent to initiate two research efforts, namely: 1) improved electrocatalysts for direct methanol electro-oxidation and 2) advanced electrolytes for methanol-air fuel cells. In pursuing these research objectives DOE should consider the synergistic interactions between electrocatalysts and the electrolyte. For example, electrocatalysts that appear to be promising in acid electrolytes are not necessarily optimum in alkaline or buffered electrolytes. The European and Japanese programs appear to have settled on acid electrolytes, whereas the small effort at Giner Inc. seems to show that buffered electrolytes have significant promise. The research program must be so structured that trade-offs between catalyst performance and electrolyte characteristics are properly considered. In addition, this research program needs to be fully coordinated with the effort initiated under the auspices of ONR and DARPA in order to ensure proper interchange of data and the minimization of duplicative efforts. Traditionally, the search for new catalysts has been Edisonian. This program should attempt to minimize such an approach and concentrate on utilizing the state-of-the-art experimental techniques to develop a predictive framework for the search of methanol oxidation catalysts.

The small investment required to initiate such an exploratory research program is certainly warranted by the potential benefits that can be obtained from the successful development of this technology. This sense was clear from the deliberations of the workshop and received the support of the various fuel cell developers and potential users present at the workshop.

The program should be reviewed in two years based on the progress of the research efforts and the conclusions of the systems analysis study. Consideration can be given at that time to broaden the scope of the program to include cell design and testing. Such an expansion in the scope of the program must depend on a thorough evaluation of the technology when compared to the goals set by the systems analysis.

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