

## FUEL CELLS FOR TRANSPORTATION

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The 150th anniversary of the Grove fuel cell is an outstanding opportunity to review the status of fuel cell technology and its potential applications. One of the most exciting and most challenging of these applications is in transportation. As shown in Fig. 1, a fuel cell powered vehicle is conceptually simple. The fuel cell provides an efficient means of converting chemical energy to electricity. If a fuel cell with adequate power capacity could be housed within the engine compartment of a vehicle, its electrical output could be used to drive an electric motor for propulsion as well as all of the electrical ancillary equipment of a modern vehicle.

In the early 1980s, Los Alamos National Laboratory developed a set of computer models and performed assessments of the performance of potential fuel cell powered vehicles [1, 2]. The conclusion of these studies was that with reasonable improvements in power density a fuel cell powered vehicle could deliver comparable performance to an internal combustion engine (ICE) powered vehicle at approximately twice the energy efficiency (*i.e.* km/l on the same fuel) and with negligible pollution. The fuel cell system could also provide comparable range and refueling time to the ICE. These have been the main deficiencies of battery powered vehicles.

The motivations to look for an alternative technology to ICEs are growing rapidly. These include: the need to improve the efficiency of fossil fuel utilization both to extend the duration of these valuable resources and to reduce the rate of carbon dioxide production; the need to reduce air and noise pollution, particularly in urban areas; and the need in many countries to reduce the dependence of transportation on petroleum.

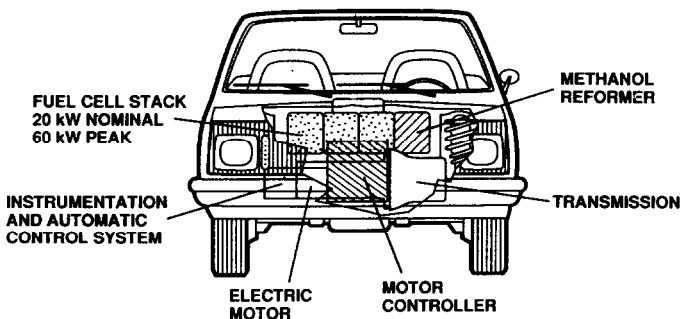


Fig. 1. Cut away view of a conceptual layout for a passenger car fuel cell propulsion system.

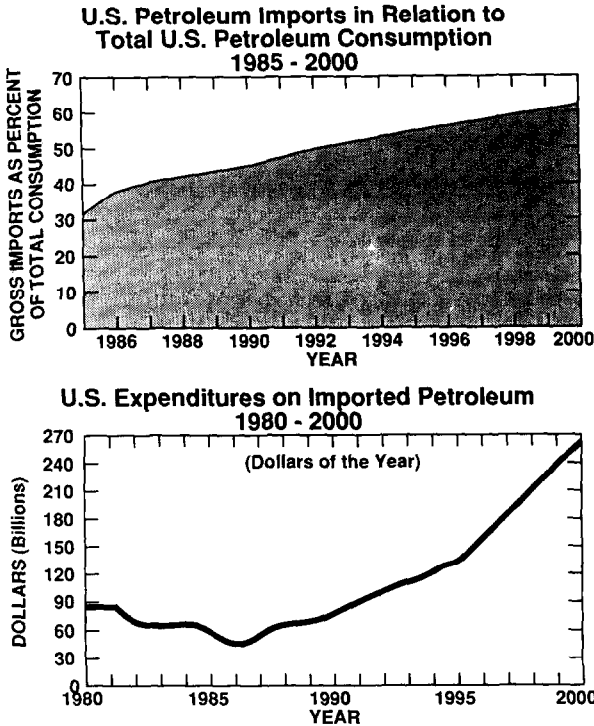
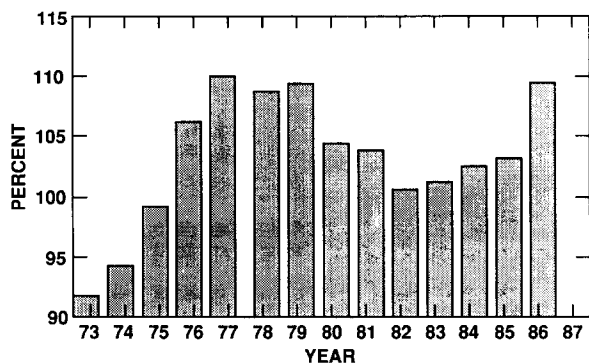


Fig. 2. The top graph shows the projected growth of United States petroleum imports in relation to total U.S. petroleum consumption. (Sources: U.S. DOE for historical and DRI Energy for projected.) The bottom graph shows the projected U.S. expenditures on imported petroleum. (Sources: U.S. Department of Commerce for historical and forecasts derived from DRI Energy projections.)

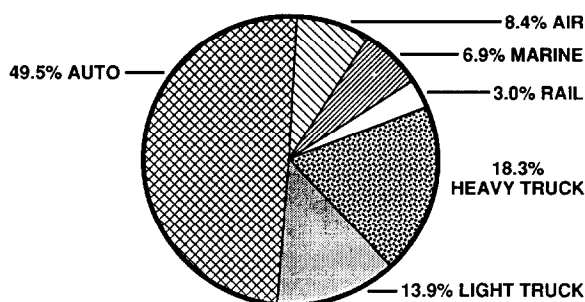
To illustrate the problem of petroleum dependence Fig. 2 shows the projected petroleum imports to the United States and their projected cost as a function of time. By the year 2000 the United States will be importing more than 60% of its petroleum at a cost of over \$260 billion per year. In the United States, most of this petroleum is used for transportation. As shown in Fig. 3, the transportation sector in the United States already requires more petroleum than is domestically produced. The bulk of this (also shown in Fig. 3) is being used in automobiles. Both the efficiency of fuel cells and their ability to use other sources of fuel could help to alleviate this petroleum dependence problem.

Pollution is also a growing problem. As shown in Fig. 4, transportation is a major contributor to  $\text{NO}_x$  pollution and many of the other components of urban smog. Fuel cells, particularly those operating at low temperature, produce negligible amounts of  $\text{NO}_x$  and much lower levels of other pollutants and particulates than internal combustion engines. They are also intrinsically quiet.



Transportation Oil Use as Percent of U.S. Oil Production

20.2 QUADS (10 MBPD) -- 1986



Petroleum Use By Transportation Mode

Fig. 3. The histogram at the top shows that transportation is a growing fraction of U.S. petroleum consumption and that this fraction has exceeded domestic production. (Source U.S. DOE.) The pie chart at the bottom shows that automobiles consume nearly 50% of the petroleum consumed by the U.S. transportation sector. (Source U.S. DOE.)

The greenhouse effect is potentially one of the most severe long term environmental problems we face. As other fossil fuels, such as coal, are substituted for petroleum this problem will worsen. Transportation is a significant contributor to  $\text{CO}_2$  production, as shown in Fig. 4. The fuel cell with its higher efficiency can reduce the rate of  $\text{CO}_2$  production in comparison with ICEs burning the same fuel.

These are among the motivations for exploring the potential of fuel cells for transportation applications. To realize this potential, major challenges must be met. The requirements for transportation applications are extremely stringent. The fuel cell must have sufficient power density to meet the performance specifications of the vehicle and to fit within the available space. It must be sufficiently inexpensive to compete with internal combustion engines on an economic basis. It must be able to start rapidly and to respond quickly to changes in power demand. It must be safe and reliable. When using hydrocarbon fuel, the fuel cell must tolerate the carbon dioxide

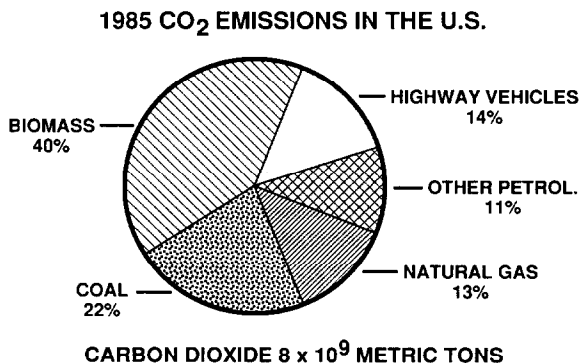
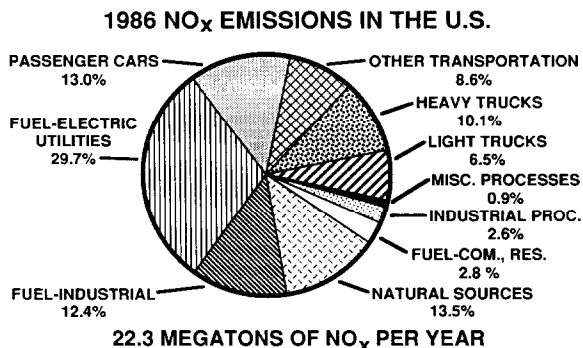


Fig. 4. The top chart shows the sources of nitrogen oxide emission in the U.S. More than 1/3 is produced by transportation. (Source: U.S. Environmental Protection Agency.) The bottom chart shows the sources of man made carbon dioxide emission in the U.S. In 1985 highway vehicles produced approximately 14%. (Source: U.S. Environmental Protection Agency.)

that is produced in the reforming process or it must be able to use the hydrocarbon directly.

All of the major types of fuel cells have been evaluated for their ability to meet these requirements. Some of the important features are summarized in Table 1. As might be anticipated, no one type is superior to the others in all respects. Indeed, one type may be more suitable than another for a specific application. For example, in ships or locomotives in which frequent on/off cycling is not required, the high-temperature fuel cells, such as solid oxide or molten carbonate, present advantages. They allow more flexibility in fuel selection and may be used without a reformer. The high-grade waste heat is more easily used in a thermally integrated system. Conversely, the low-temperature fuel cells, such as polymer electrolyte or alkaline, may be a better choice for passenger cars to which rapid start-up and wide power range are important.

TABLE 1

## Fuel cell comparison

Fuel cell type	Operating temperature (°C)	CO <sub>2</sub> tolerant	Power density H <sub>2</sub> -air (W/cm <sup>2</sup> )	CO tolerance	Reformer required
Solid oxide	1000	yes	0.20 - 0.27	good	no
Molten carbonate	600	yes	0.19 - 0.24	good	no
Phosphoric acid	150 - 205	yes	0.20 - 0.29	fair	yes
Alkaline	65 - 220	no	0.15 - (4.3)	poor	no
Solid polymer	25 - 120	yes	0.2 - 0.9	poor	yes

Recently, major advances have been achieved in fuel cell technology, which make the application to transportation much more viable. These advances include large increases in power density, major reductions in intrinsic cost and improved system design.

For example, Argonne National Laboratory in the United States has been developing a monolithic solid oxide fuel cell that has the potential for very high power density [3]. These cells operate at  $\sim 1000$  °C, potentially eliminating the need for a reformer. The monolithic design employs the same materials used in the tubular solid oxide design. These materials provide excellent thermal-expansion matching, which is critical for cycling to the operating temperature of the cell. For the monolithic cell, the materials are deposited by tape-casting techniques. Because the layers are thin, high specific power is potentially available. Current work is focused on improving the fabrication procedures, improving the materials integrity, and on reducing interface resistances in stacks.

Much interest in alkaline fuel cells for transportation has been evident in Europe, where hydrogen is more widely available. Although it cannot withstand CO<sub>2</sub> in the fuel stream, the alkaline fuel cell offers very high power density and efficiency. In Belgium, Elenco has developed a 15 kW alkaline fuel cell for powering an electric test van [4]. This company is particularly interested in commercially operated vehicles (city buses, refuse trucks, vans, etc.) for transporting people or goods in urban areas.

At Los Alamos National Laboratory with support from the United States Department of Energy, we have focused our attention on the polymer electrolyte fuel cell. This technology offers a combination of characteristics, which we feel make it the leading contender for passenger vehicles. It provides high power densities with values over 2 W/cm<sup>2</sup> on H<sub>2</sub> and O<sub>2</sub> as reported by Ballard Technologies Corporation in Canada [5]. It can tolerate CO<sub>2</sub> in the fuel stream thus allowing the use of reformed hydrocarbon fuel. It can be self-starting at temperatures above  $\sim 20$  °C. Low-cost structural materials can be used because of the low operating temperature and reduced corrosion. And the solid character of the electrolyte facilitates sealing and safety of the fuel cell stack.

**DEVICE PRODUCES ELECTRICAL ENERGY FROM  
HYDROGEN OXIDATION**

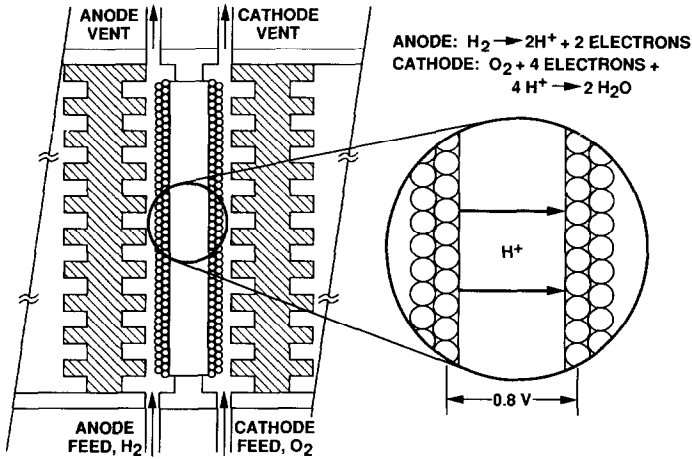
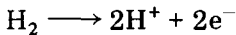


Fig. 5. Schematic cross section of a polymer electrolyte fuel cell.

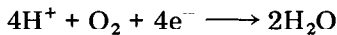
The limitations of the technology have been high platinum content, expensive membranes, poor CO tolerance, water management problems and difficulty in thermally integrating with a reformer. The objective of the Los Alamos program has been to solve these problems.

The cross section of a single polymer electrolyte fuel cell is shown schematically in Fig. 5. Typically the cell consists of graphite bipolar plates which are pressed against the membrane-electrode assembly. These plates have a manifold of grooves which distribute the reactant gases to the electrodes. They are also sufficiently electrically conductive to pass the generated current to the adjacent cell.

The membrane-electrode assembly is the electrochemical heart of the system. On the anode or hydrogen side, hydrogen gas is catalytically dissociated according to the reaction



The hydrogen ions pass through the polymer electrolyte to the cathode or oxygen side of the cell. There they are combined catalytically with oxygen and electrons from the adjacent cell to form water, according to the reaction



The polymer electrolyte, from which the cell derives its name, is a remarkable ionically-conducting, plastic-like material in the form of a membrane 50 to 175  $\mu\text{m}$  thick. Membranes sold commercially by DuPont under the Nafion<sup>®</sup> trade name and several types of experimental membrane manufactured by DuPont, Dow and other companies are used. These materials are perfluorosulfonic acids (teflon-like fluorocarbon polymers with side chains ending in sulfonic acid groups). To maintain their protonic conductivity the

membrane must contain sufficient water. In operation, maintaining the correct water content is one of the key aspects of polymer electrolyte fuel cell design.

In order for the electrochemical reactions to take place at useful efficiency they must be catalyzed. To date, platinum has proven to be the best catalyst for both the hydrogen oxidation (anode) and the oxygen reduction (cathode) reactions. To function, the catalyst must have access to the gas and must be in contact with both the electrical and protonic conductors.

This may be achieved by pressing the platinum catalyst into the membrane and then contacting it with a porous electrode that provides both electrons and gas access. The deficiency of this approach is that relatively large amounts of platinum must be used to obtain adequate catalytic activity. Typically, 4 mg of platinum are used on each square centimeter of electrode area. The problem is that platinum is much too expensive to be used in this quantity for a consumer application. With such loading, an automobile would require up to \$10 000 worth of platinum.

To solve this problem, at Los Alamos the use of supported platinum catalysts similar to those used in liquid electrolyte fuel cells is being investigated. The supported catalyst consists of 2 to 5 nm diameter Pt particles on the surface of fine carbon particles. This greatly increases the effective surface area of the platinum. In much of this work electrodes manufactured by Prototech Inc. in which these carbon particles are mixed with teflon and bonded to a porous carbon cloth have been used. Unfortunately when such electrodes are pressed against the polymer electrolyte membrane only those few catalyst particles at the surface make effective contact with the protonic conductor.

This problem has been solved by impregnating the supported catalyst electrode with protonic conducting material. This is achieved by covering the surface with a solution of solubilized Nafion<sup>®</sup>. As shown schematically in Fig. 6, if the solution is the appropriate concentration, it will leave a

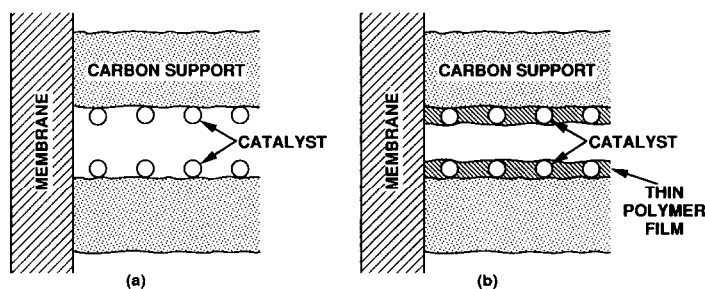


Fig. 6. Schematic representation of the technique developed at Los Alamos for use of supported platinum catalysts in polymer electrolyte fuel cells. In (a) the catalyst particles are ineffective because protons from the membrane cannot migrate across the carbon surface. By impregnating the pores of the electrode with a thin layer of protonic conductor (b) the supported catalyst becomes highly effective.

protonically conducting film on the catalyst particles that connects them to the membrane without blocking gas access. This technique has proven to be extremely effective. Performance comparable to the high catalyst loading cells has been achieved with approximately 1/20 the platinum, reducing the catalyst cost for an automobile sized fuel cell to a few hundred dollars.

The performance characteristic of a fuel cell is shown schematically in Fig. 7. As the current drawn from the fuel cell increases the voltage falls. Ideally at low currents the voltage would be near the reversible potential, 1.23 V. In practice, the catalyst does not function perfectly and produces activation losses of 0.2 to 0.3 V. At higher currents, ohmic losses due to the finite resistance of the cell contribute an additional 0.1 to 0.3 V of loss. At sufficiently high current densities (1 to 4 A/cm<sup>2</sup>) reactant gases can no longer be supplied to the catalyst sites at sufficient rates to sustain the reaction and the voltage drops rapidly to zero. In this regime the power output from the cell begins to fall. Each of these voltage losses also corresponds to a loss of efficiency, since the efficiency is simply the ratio of the cell voltage to 1.48 V.

For comparison, Fig. 7 shows the current–voltage characteristic of a polymer electrolyte fuel cell using low catalyst loading electrodes. This cell

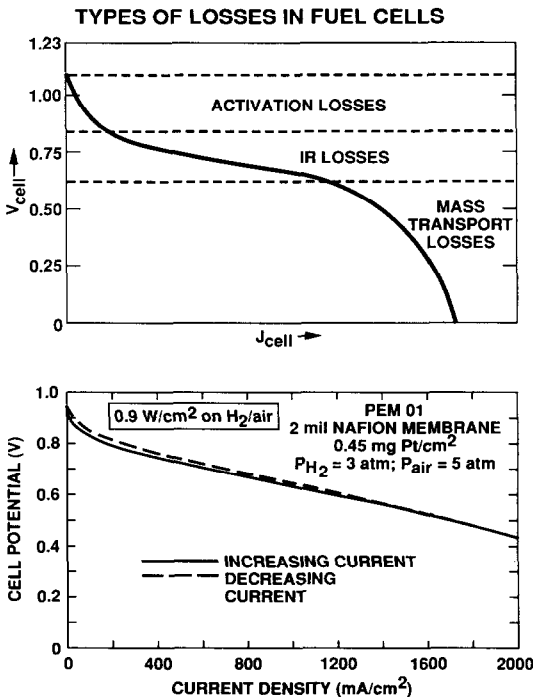


Fig. 7. The top graph is a conceptual representation of the current–voltage characteristic of a fuel cell, showing the major contributors to voltage loss. The bottom graph shows an experimentally measured current–voltage characteristic of a polymer electrolyte fuel cell using low platinum loading electrodes.



TABLE 2

Fuel cell parameters for a passenger car

Net continuous power	20 kW		
Net peak power	60 kW		
Operating points			
Peak	0.50 V	1.8 A/cm <sup>2</sup>	0.9 W/cm <sup>2</sup>
Continuous	0.75 V	0.4 A/cm <sup>2</sup>	0.3 W/cm <sup>2</sup>
Stack size			
Active area	500 cm <sup>2</sup>		Diameter 25 cm
Cross section	1000 cm <sup>2</sup>		Diameter 35 cm
Cell thickness	0.5 cm		
No. cells	133		
Stack length	66 cm		Total 75 cm
Stack voltage (nom.)	100 V		
Stack volume	0.072 m <sup>3</sup>		
Stack density	1.0 g/cm <sup>3</sup>		
Stack weight	75 kg		Total 100 kg

TABLE 3

Comparison of hydrogen storage technologies

Technique		cal/g	cal/cm <sup>3</sup>
Gas	steel cylinder (200 atm)	510	240
Liquid	cryogenic H <sub>2</sub>	4250	2373
Hydrocarbon	n-octane	11400	8020
	methanol	5340	4226

is capable of delivering a power density of 0.9 W/cm<sup>2</sup> at a current density of 2 A/cm<sup>2</sup> using hydrogen and air. A practical power plant is made by stacking a large number of such cells in series. Table 2 shows a set of design parameters for a stack of 133 such cells with an active area of 500 cm<sup>2</sup> each. The full stack would provide 100 V at 200 A at the continuous operating point of 20 kW. The power output could be increased to 60 kW by increasing the current density to 1.8 A/cm<sup>2</sup>. With individual cell thicknesses of 0.5 cm the stack would be approximately 75 cm long and 35 cm in diameter with a weight less than 100 kg including assembly hardware and pressure housing. Such a stack could easily meet the power density requirements for a compact car giving performance comparable to that provided by a standard 4 cylinder ICE power plant.

The choice of fuel is one of the key factors in the design of a fuel cell propulsion system. Most fuel cells operate best on pure hydrogen and oxygen. Usually air can be used directly as the oxidant with little loss of performance. Pure hydrogen can be carried aboard a vehicle either as compressed gas or as a cryogenic liquid. However, as shown in Table 3, the

volumetric energy density of pressurized hydrogen is poor. The energy density of liquid hydrogen, though better, is still less than 1/3 that of gasoline, and the technology for storing small volumes of liquid hydrogen is not well developed. Hydrocarbons by comparison provide a simple way to store hydrogen at high density. In some types of fuel cell, hydrocarbons can be used directly. In others they must be chemically converted to hydrogen by a catalytic process called reforming. As a reformed fuel, methanol is particularly attractive. It can be efficiently converted to  $H_2$  and  $CO_2$  by reacting it with water on a Cu-Zn catalyst at  $\sim 180^\circ C$ .

In the United States particularly, methanol is a likely fuel to replace gasoline in the transportation sector as petroleum reserves are depleted. Methanol can be synthesized from a variety of domestic resources including coal, natural gas and biomass. It is a liquid fuel that is easily integrated into the existing distribution system, and has about half the energy density of gasoline. Since a fuel cell propulsion system has about twice the energy efficiency of an internal combustion engine, it can achieve comparable range on a tank of methanol to that of an ICE burning gasoline from a tank of the same size.

To use methanol in a polymer electrolyte fuel cell propulsion system, one must have an on-board reformer that can meet the vehicle requirements. These include rapid start-up and quick transient response to meet the changing fuel demands. Conventionally reformers have not been designed to meet these requirements, often taking an hour or more to start up and many minutes to change output significantly.

At Los Alamos reformer designs which may solve these problems have been investigated [6]. One approach is shown schematically in Fig. 8. In

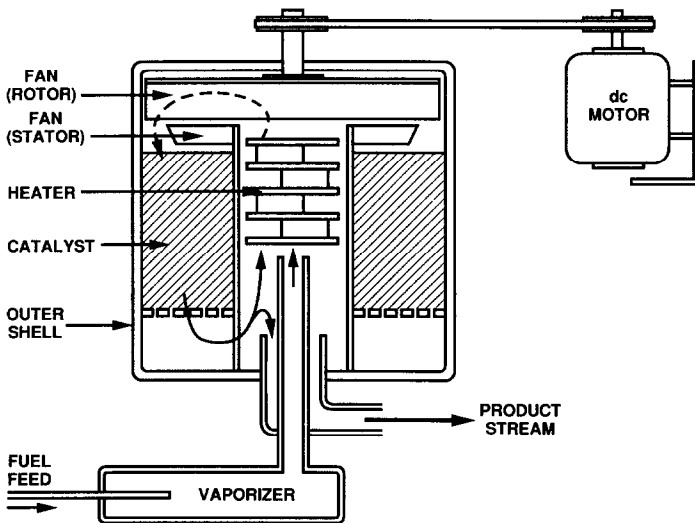


Fig. 8. Schematic cross section of a recirculating methanol-water reformer developed at Los Alamos.

this design an internal fan is used to recirculate the reformat through the catalyst bed. In this way, the catalyst bed can be kept uniformly at its optimal temperature, and heat can be rapidly injected to increase the reforming rate.

One of the problems with methanol-water reformers is that the reaction is often not complete, leaving small concentrations (approximately 1%) of carbon monoxide in the fuel stream. This carbon monoxide is an extremely effective poison to the fuel cell catalyst. As shown in Fig. 9, even a few parts per million CO produce a substantial degradation in the fuel cell performance, particularly at high current densities.

One approach to solve this problem is to reduce the carbon monoxide content of the reformat. This has been achieved by selectively oxidizing the CO to  $\text{CO}_2$  [5, 6]. To achieve this, the reformat passes through a small reactor containing a platinum catalyst. As shown in Fig. 10, by injecting a small amount of oxygen or air into this reactor, the CO content can be dramatically reduced with relatively little hydrogen consumption.

At Los Alamos, recent work has shown that the performance degradation caused by trace amounts of CO still remaining in the reformat can be restored by injecting small amounts of air at the anode [7]. For example, Fig. 11 shows that the severe performance degradation caused by 100 ppm CO can be completely restored by injecting 2% air into the anode feed.

Much has been accomplished over the last few years in solving some of the key problems that impeded the use of fuel cells in transportation. The polymer electrolyte fuel cells in particular are looking increasingly attractive for this application. The power density has been greatly increased, the platinum requirements have been dramatically reduced, and solutions to carbon monoxide poisoning have been found. Much work still remains to be done before this technology can become commercial. However, the viability

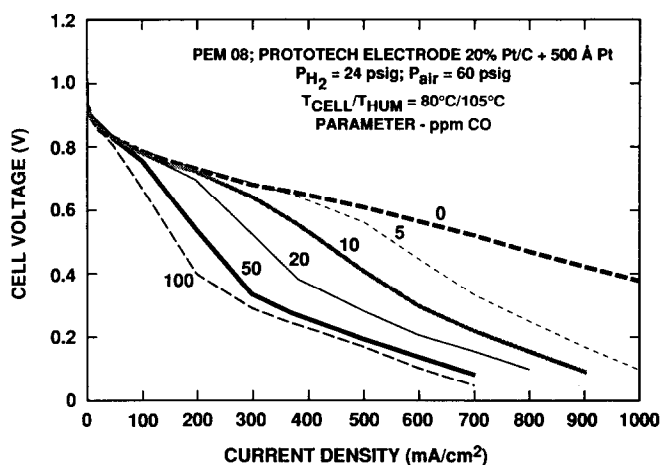


Fig. 9. This graph shows the performance losses caused by trace amounts of carbon monoxide in the fuel stream.

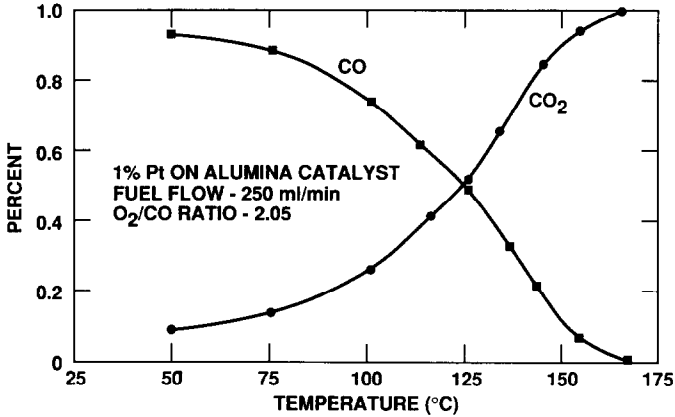


Fig. 10. This graph shows that the residual carbon monoxide in the methanol reformat can be effectively converted to carbon dioxide by post-reformer selective oxidation at a temperature of  $\sim 160$  °C.

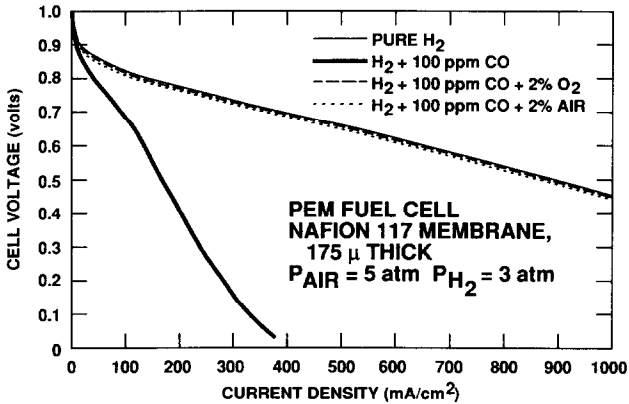


Fig. 11. This graph shows that residual catalyst poisoning by trace amounts of CO can be completely eliminated by injecting  $\sim 2\%$  air into the fuel cell anode.

of the technology is sufficiently well established to begin to solve the problems of system integration.

From the system perspective, the fuel cell stack is just one of a large number of subsystems that must be integrated to produce an effective propulsion system. The complexity of this task is indicated in Fig. 12.

These difficult problems are beginning to be addressed. The U.S. Department of Energy (DOE) and the U.S. Department of Transportation (DOT) have initiated a cooperative multiyear program with industry for the research, development and demonstration of a fuel cell/battery hybrid bus system for urban passenger transport [8]. In its early development, this system will be based upon a phosphoric acid fuel cell (PAFC), sized to provide the average power requirements of the bus, and a battery pack

## FUEL CELL PROPULSION SYSTEM

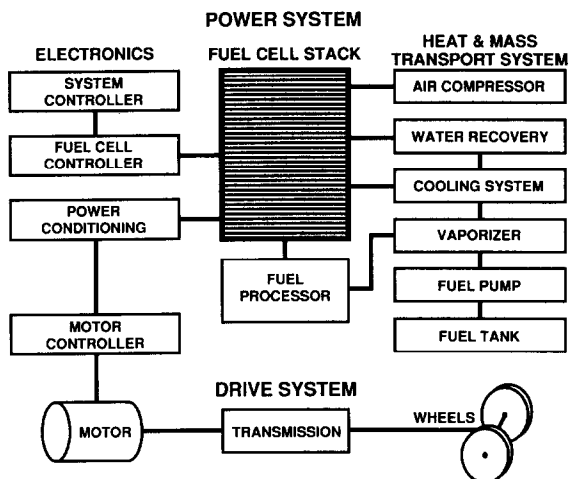


Fig. 12. Block diagram of a fuel cell propulsion system showing the large number of subsystems that must be integrated.

capable of meeting the peaking requirements [9]. The PAFC system was selected for the early phases of this project because it is currently the most highly developed fuel cell system. It is expected that the polymer electrolyte fuel cell will replace the PAFC in the latter stages of the program.

In conclusion, fuel cells offer an attractive alternative power source for transportation applications. They provide over twice the efficiency of internal combustion engines and can operate on non-petroleum-based fuel, such as methanol. Significant advances have been made in increasing the power density and reducing the intrinsic cost of fuel cells, but much work remains before they can be successfully integrated into vehicles on a large scale.

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