

Fuel Cells: a New, Efficient and Cleaner Power Source

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Introduction

Fuel cells are in the spotlight. However, they are anything but new. They are older than the internal combustion engine, invented by Nikolaus August Otto in 1876, and the compression/ignition engine, invented by Rudolf Diesel in 1892. The fuel cell or “gas battery” was invented in the 1840s by Sir William Robert Grove, but the discovery of the “fuel cell effect” by professor Christian Friedrich Schoenbein dates back to 1838.

Several factors, such as the success of the internal combustion engine and associated petrol industry, had contributed to its slow development until a resurgence of interest during the Gemini space program in the 1960s due to its multiple inherent advantages, such as reactant commonality with rocket engine fuel and oxidant. Since that time, developments in the field have been relatively rapid and continuous, although often erratic. The reasons behind the renewed interest are well documented (Kordesch and Simader, 1996; Hirschenhofer et al., 1998), which include these advantages over conventional power sources: high energy efficiency, high energy and power density, environmental friendliness (low pollutant emissions and absence of radiation), low noise, rapid refueling, and fuel flexibility. For special applications, such as aerospace, additional advantages are present: e.g., heat production and potable water generation. In this article, the state of the art of fuel cells, as well as manufacturing and research challenges, will be reviewed.

Status of fuel cells

A fuel cell is an electrochemical energy conversion device, which is similar to, but different from, a battery. Whereas a battery stores the energy in the chemicals contained in it, a fuel cell acts as a converter—the reactants and products merely being in transit in the unit (battery electrodes are consumed, while fuel cell electrodes are not). A typical fuel cell with a hydrogen fuel and air as an oxidant is illustrated in Figure 1. At the anode, a fuel (H_2 , CH_4 , or CH_3OH) is oxidized, whereas, at the cathode, an oxidant (O_2 in air, O_2 , CO_2) is reduced. In this spontaneous process, electrons generated at one electrode and consumed at the other circulate in an external circuit which can drive, for example, an electric motor.

A number of different subsystems are needed to allow the fuel cell to properly function. These additional system components include: oxidant and fuel delivery sub-systems; a cooling subsystem to manage the heat generated; a control subsystem to ensure

harmonious interactions among all these fuel cell system components; and a power conditioning sub-system such as a dc-to-dc or dc-to-ac converter. In addition, proper design of these subsystems, as well as the fuel cell stack, is required to ensure overall maximum efficiency and reliability. For example, the use of diluted reactants and/or the formation of a product affects mass transfer in a significant manner, which requires careful engineering consideration. Similarly, the presence of a distributed current in the fuel cell reactor is a major consideration, as the local current density can significantly affect material properties and degradation, and performance and reliability, if areas of high current density are present (hot spots), or the current density distribution is not uniform (efficiency loss). Fuel cell reliability is also affected by the presence of contaminants (St-Pierre et al., 2000a) in the reactant streams which may be present at its source, are created within the fuel cell system (e.g., in the fuel reformer), or are transported from component materials by the action of the reactant streams on the fuel cell stack. Knowledge of all these aspects (and more) is required before optimization and final design can be attempted.

The fuel cell system design also depends largely on the fuel cell type. Fuel cells are classified commonly by the electrolyte being used (alkaline, polymer, phosphoric acid, molten carbonate, and solid oxide) or by their operating temperatures ($\sim 80^\circ\text{C}$ for alkaline and polymer; $\sim 200^\circ\text{C}$ for phosphoric; ~ 600 – 650°C for molten carbonate; and ~ 800 – $1,000^\circ\text{C}$ for solid oxide). Biological fuel cells, which are comparatively still in their infancy, will not be discussed here (Palmore and Kim, 1999). For high-temperature fuel cells (molten carbonate and solid oxide), mass-transfer considerations may be ignored due to higher reactant and product diffusivities. In addition, the elevated temperatures allow a combination of the reformer with the fuel cell stack (the anode catalyst and/or electrolyte combine reforming and electrocatalyst functions).

Such design features are beneficial since they can reduce fuel-cell system's complexity and increase reliability. However, dynamic operation issues including startup are more pronounced for such fuel cells, and, as a consequence, they are generally considered essential for stationary, continuous power applications (> 100 MW). Similar dynamic operation, as well as other considerations, have dictated the use of phosphoric acid fuel cells for small stationary applications (< 10 MW), alkaline fuel cells for aerospace applications (CO_2 contaminant in the reactant streams cannot be tolerated by the electrolyte), and polymer electrolyte fuel cells for micro, portable, motive and small stationary applications.

Current fuel-cell systems can achieve efficiencies from 40 to 60%, and even up to 80%, if high-grade heat is reused. These val-

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ues compare favorably to the $\leq 20\%$ achieved by internal combustion engines or to the 50% achievable by gas turbines. With respect to energy density, fuel-cell stack values up to 3 kW/l have been either demonstrated or claimed. *Fuel cells also emit significantly less pollutants.* This is the result of a number of factors including high energy efficiency (need less fuel to achieve the same amount

of work), low operating temperature (generation of NO_x is only favored at high temperatures), and the possibility of using fuels such as renewable hydrogen and methanol. Of course, any pollution claim needs to be examined in a global and life cycle context since, for example, the impact of the fuel generation and fuel-cell component production technologies on the environment could be

Polymer Electrolyte and Phosphoric Acid Fuel Cells

Figure 1a

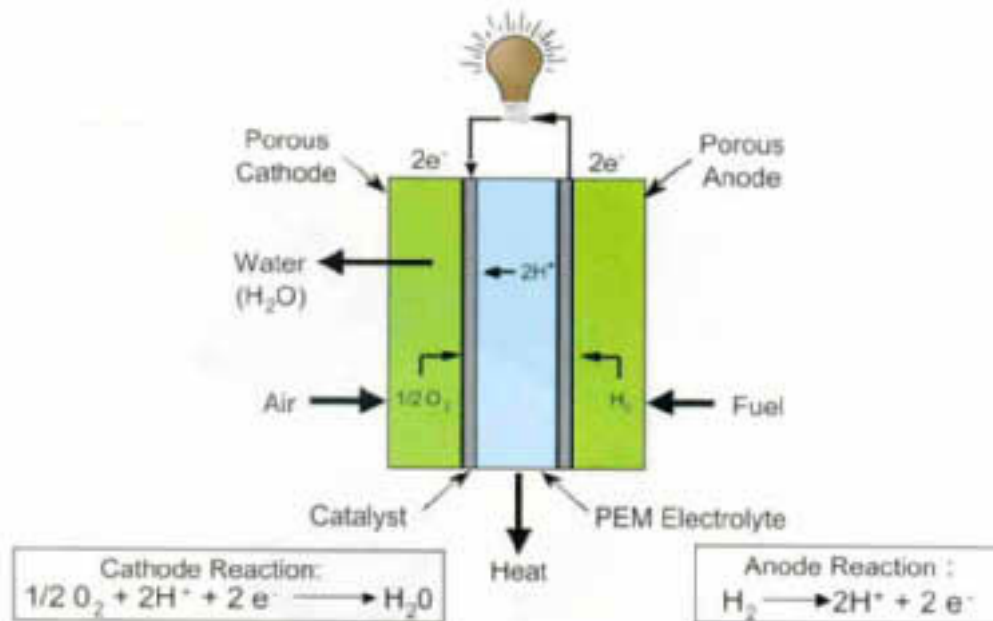


Figure 1b

A Polymer Electrolyte Fuel Cell Stack

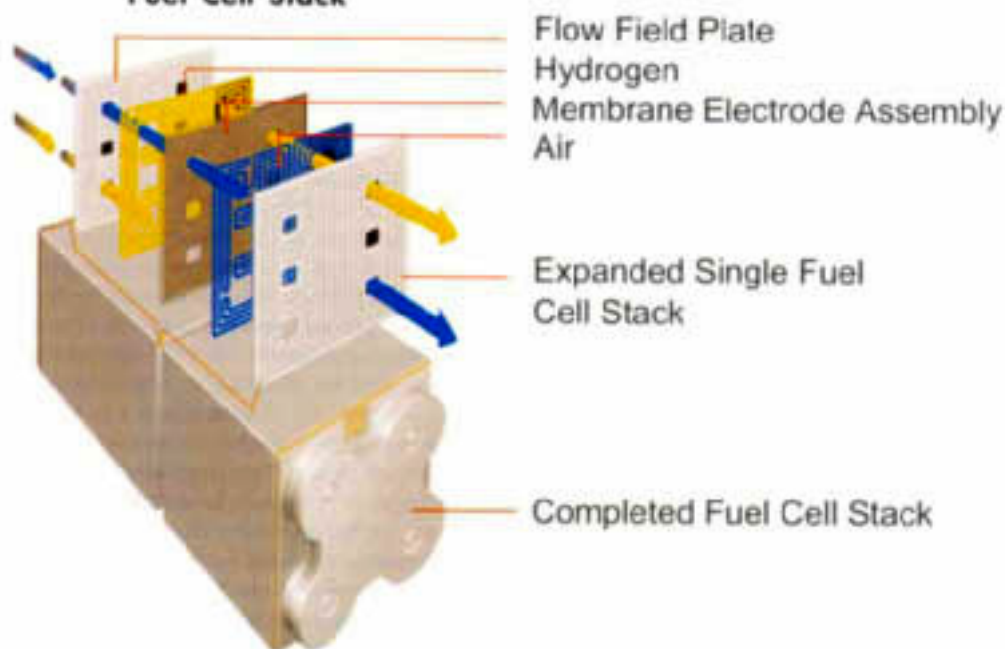


Figure 1(a) Generic fuel cell (polymer electrolyte or phosphoric acid); (b) typical fuel cell stack with oxidant and fuel reactant flowing through internal manifolds and flow field plates.

an issue. Since fuel cells do not have moving parts, noise levels achieved by systems are low with the majority of noise coming from such subsystems as an air compressor. Refueling for fuel cells, although requiring upgrades to existing gas stations to adapt to different fuels, does not require significantly more time than with gasoline or diesel. Finally, many fuels can be used to power fuel cells. In addition to those already listed, propane, butane, and hydrogen stored in metal or other hydrides such as liquid NaBH_4 represent other alternatives ($\text{NaBH}_4 + 2 \text{H}_2\text{O} \rightarrow 4 \text{H}_2 + \text{NaBO}_2$), (Amendola et al., 2000).

Future engineering challenges and prospects

There has been an increased interest in fuel cells for a number of different applications (micro, portable, stationary, motive, marine, and aerospace) with a rapid rise in markets. For example, annual growth rate of 30% has been predicted for stationary applications with sales expected to reach \$1.4 billion by 2003 (Weidner and Doyle, 2000). Several hundred companies around the globe are involved in the field. Since the technical feasibility of fuel cells has been to a large extent successfully demonstrated, the industry is currently engaged in field trials, lowering costs, and determining manufacturing strategies and processes for these fuel-cell systems. Manufacturing challenges include maintaining fuel-cell performance, introducing low-cost materials, implementing high-volume methods with high yields, simplifying fuel-cell design (reduction in part number), and establishing supplier relationships (economy of scale). Technical advances and breakthroughs are still required to help further increase reliability and address fundamental issues at the same time, e.g., by simplifying the fuel cell system (component elimination), reducing heat rejection at high current densities, improving reactant flow distribution within stacks, designing stacks less prone to failure (by decreasing the number of cell in series), or by integrating new technologies from other fields of science that would lead to net overall system benefits.

This is especially true for motive applications since requirements are the most stringent (lowest size and cost). The challenges are best illustrated by considering recent advances (Figure 2). For example, the water vapor space velocity for polymer electrolyte fuel cells has increased from 1.6 to 15.7 $\text{cm}^3/\text{cm}^3 \text{ s}$ between 1991 and 1998. This is a large value for these systems considering that approximately 1/3 of the volume is associated with each component, the anode, cathode and coolant flow fields, and that part of the volume is required to provide structural support, as well as separating the different streams. Therefore, the usable amount of space available to circulate the streams is relatively small and creates significant challenges

to chemical engineers. A similar situation exists for heat transfer by considering the advances in power density, which resulted in an increase in heat rejection from approximately 0.35 to 3.4 W/cm^3 .

System Design. The selection or design of a fuel-cell system depends to a large extent on the application. For example, for portable applications, it is preferable to eliminate the air compressor and use a fan to simplify the system. Such a selection further impacts system design since the operating pressure is low in this case (< 1.3 bara). Another example relates to fuel cells operating with a reformat fuel. Typically, such a system would require a fuel reformer and a number of unit operations to remove contaminants that can affect downstream steps. For the reforming step, thermal decomposition, steam reforming, partial oxidation, and autothermal reforming technologies are available. For the cleanup steps, water/gas shift reactors, selective oxidation, membrane separation, and pressure swing

adsorption technologies could be used. For low temperature fuel cells, Pt group metals are currently used for the anode electrocatalyst. As a consequence, the CO level in the reformat has to be very small (e.g., < 10 ppm) to minimize the performance drop associated with CO adsorption. This is not a critical consideration for higher-temperature fuel cells, such as phosphoric acid, some of which can reform fuels directly inside the stack (Park et al., 1999).

Many fuel cell systems have been proposed and some demonstrated, but the best combination of system components has not yet emerged due in part to the uncertainties related to fuel distribution (e.g., the methanol and hydrogen infrastructure is not well defined), availability of fuel cell purpose

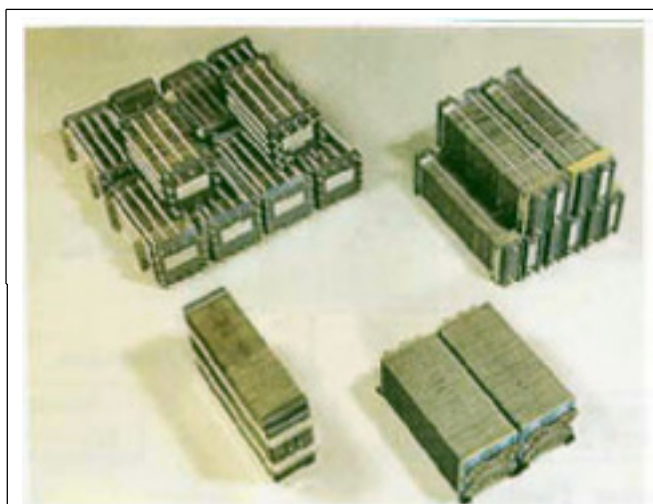


Figure 2. Number of Ballard polymer electrolyte fuel-cell stacks required to provide 50 kW with increasing power density.
Moving clockwise from the upper left corner, Mk300 (1991), Mk500 (1993), Mk700 (1995), Mk800 (1997).

built components (compressors, valves, compact reformers, etc.), and absence of suitable models and data for optimization. However, some trends can still be identified with a relative degree of certainty such as the need for system simplification, and increased power density and reliability. New components such as highly efficient and compact pressure swing adsorption devices have recently been developed (e.g., up to 2,500 $\text{l O}_2/\text{l adsorbent h}$) (Keefer et al., 2000), which offer the possibility of increasing fuel-cell performance by increasing inlet O_2 concentration and minimizing mass-transfer losses, as well as considerably simplifying the fuel reforming process.

Reactor Design. The selection of a reactor design for a fuel cell stack/system is similarly constrained by the application envisaged. Many reactor types have already been studied such as the plug-flow reactor, the batch reactor, the continuous stirred-tank reactor, and their modifications. The plug-flow reactor is the most popular configuration (spent air and reformat are fed to a catalytic burner after passing through the stack to provide heat to the reformer and minimize emissions). The plug-flow reactor with recycle is useful particularly for pure reactants (O_2 , H_2), which are more costly to synthesize than readily available reactants such as air or

methanol/water fuel solutions from which product CO_2 is easy to separate. The plug-flow reactor with distributed feed was also suggested as a method to help minimize the CO formation in a reformat fuel stream by the reverse water gas shift reaction ($\text{CO} + \text{H}_2\text{O} \leftrightarrow \text{H}_2 + \text{CO}_2$) by carrying the CO oxidation step as close as possible to the anode chamber and therefore minimizing the residence time and production of CO (Wilkinson et al., 1996).

The batch reactor is convenient especially for portable applications due to fuel storage restrictions, whereas the continuous stirred tank reactor behavior is approached by fuel cells considered for aerospace applications (O_2/H_2 feed). Parallel flow distribution reactors are mainly used (all cells in a stack are fed with reactant at the same time). In some cases, however, cascaded flow distribution reactors where groups of cells in a stack are fed in series (Winsel, 1971) are used in cases where reactant stoichiometries need to be low if recirculation needs to be avoided for simplification (e.g., with H_2 fuel in a portable application). Other reactor configurations can benefit fuel cell systems, especially to address specific issues or to take advantage of new opportunities. As already mentioned, the advent of highly efficient pressure swing adsorption systems increasing reactant concentrations offers a path to reduce mass transport issues especially for the oxidant stream. New reactor designs or modifications of existing ones would ensure that fuel cell efficiency is optimized by taking advantage of the more concentrated reactant streams that can be produced. This is yet another challenge for fuel-cell reactor design.

Mass Transfer. Mass transfer plays a significant role in reactor design, especially for low temperature fuel cells, as explained previously. This is due to the existence of significant gradients in concentration and temperature, which are the result of electrochemical reactions, and to system related requirements (reactant stoichiometries and coolant flow rates required to be low to reduce parasitic power), which further complicates issues by creating nonuniformities. For example, in polymer electrolyte fuel cells, water is required for the ion-exchange polymer to be conductive. In systems operating without external reactant humidification (for simplification), this creates a weak area at fuel-cell inlets due to the resulting dry region. The fuel-cell outlet can, however, contain a substantial amount of water, which creates a mass-transfer problem. This complication is further affected by the decrease in reactant concentration from the inlet to the outlet. Several methods are available to mitigate these issues and include reactor design, flow field design, electrode design, and operating conditions selection (Wilkinson and St-Pierre, 2001). For example, the electrode porosity can be changed to accommodate changes occurring along the flow field length. Additionally, operating conditions can be manipulated by allowing the temperature to sufficiently increase near the outlet to allow evaporation of the water produced and/or by condensing water in a dry region by lowering the temperature. Managing mass-transfer issues will continue to be a very challenging area of development considering the continuous demands on increased power density (smaller fuel cells) as well as the potential of new technologies that could significantly impact design activities.

Heat Transfer. In comparison to mass-transfer issues, heat transfer has not received the same amount of attention from fuel-cell developers and relatively little has been published in this area. Both liquid-phase [in a separate flow field or mixed with reactants (Wilkinson et al., 1999), liquid electrolyte] and gas phase coolants (reactants) have been used to manage heat generated within fuel cells. The coolant selection is, of course, partly dictated by the fuel-

cell type, especially the fuel cell temperature with liquid coolants generally being more appropriate for low-temperature fuel cells (greater need to remove heat to keep the operating temperature low) and gaseous coolants being preferred for high-temperature fuel cells (heat maintains the high operating temperature, whereas reactant circulation fine-tunes it to the desired level and uniformity). The method used to distribute coolants is also flexible depending on the temperature distribution required. The cell/stack design can be adapted to the heat flux in the fuel-cell plane (Wozniczka et al., 1998) and/or perpendicularly to that plane. For liquid coolant selection, different factors play a role: availability, cost, physical properties (such as thermal and electrical conductivity, heat capacity, viscosity, freezing point, ion solubility, etc.), and contamination risk to fuel-cell system components. Automotive coolants, such as the well-known water/ethylene glycol system, have used inhibitors to limit degradation which poison the fuel cell (ethylene glycol degrades with time due to an autocatalytic reaction). However, for low-temperature fuel cells, the operating temperature is significantly lower than in an internal combustion engine, which significantly reduces glycol decomposition rates. Further reduction in decomposition rates is achieved by adding an ion-exchange resin to capture the products of decomposition (St-Pierre et al., 2000b). This system has the added advantage of maintaining the ionic conductivity of the solution at a sufficiently low level and, therefore, minimizes the risks of stray currents. These currents arise in the presence of conductive coolants, which are subject to potential differences locally creating material degradation by corrosion and decreasing fuel cell efficiency. This field could benefit from further study in view of glycol toxicity and other concerns.

Current Distribution. Current distribution is a design parameter, which is not generally a consideration for most reactors, but which adds a new layer of complexity to electrochemical engineering processes. This is an important area, which has also been somewhat neglected, although it may significantly impact fuel-cell efficiency, as well as reliability. Uneven current distribution is the result of nonuniformities in electrolyte conductivity, kinetic and mass-transfer limitations, and reactant concentration. This can result in hot spots (areas of higher than average current density) creating mechanical/thermal stresses that can negatively affect fuel-cell materials eventually leading to failure. An uneven current distribution also results in a lower fuel-cell efficiency since areas of lower current density do not contribute as much to the production of power. Although a number of methods are available to diagnose current density distributions (Stumper et al., 1998), such as masking sections of an electrode active area, separating the active area of a fuel cell into several independently controlled units, or using a segmented flow field plate, few in-depth studies are available. Current distribution is still an important aspect of fuel cell design.

Conclusion

The rise in pollution (regulations are becoming more stringent), fossil fuel reserve limitations, and deregulation of the distributed power sector (driven by power losses, electromagnetic radiation hazards, plant siting, and plant cost concerns) are concerns that have and will continue to increase global concerns. These issues represent strong driving forces behind the development of new power sources with potentially zero-emission fuel life cycles. Hydrogen as a fuel has the potential to be a zero emission life cycle energy carrier, as long as it is produced by renewable technologies, such as hydroelec-

tric, solar, geothermal, wind and biomass (long-term sustainability). However, a substantial amount of work is still required to achieve such a vision since hydrogen still has issues (fuel infrastructure and storage) and < 2% of the world energy is produced from renewable sources. Irrespective of the fuel selection, fuel cells are a real alternative for energy conversion in the near and long-term future. In a relatively short time frame, a number of organizations around the globe have taken on the challenge to commercialize fuel cells. Some key engineering aspects of fuel cells have been discussed, but many other areas, such as modeling, reliability, catalysis, sensors, manufacturing, and control, represent additional challenges awaiting to be addressed to bring fuel cells to a full commercial scale. Chemical engineering can play a major role in this endeavor, and many present and future career opportunities exist in various fields, especially in the areas of electrochemical and chemical engineering.

Literature cited

- Amendola, S. C. S. C., S. L. Sharp-Goldman, M. Saleem Janjua, M. T. Kelly, P. J. Petillo, and M. Binder, "An Ultrasafe Hydrogen Generator: Aqueous Alkaline Borohydride Solutions and Ru Catalyst," *J. Power Sources*, **85**, 186 (2000).
- Hirschenhofer, J. H., D. B. Stauffer, R. R. Engleman, and M. G. Klett, *Fuel Cell Handbook*, 4th ed., Parsons Corporation (1998).
- Keefer, B. G., C. Mclean, and M. J. Brown, "Electrical Current Generation System," PCT Application WO 00/16425 (Mar. 23, 2000).
- Kordesch, K., and G. Simader, *Fuel Cells and their Applications*, VCH (1996).
- Palmore, G. T. R., and H.-H. Kim, "Electro-Enzymatic Reduction of Dioxygen to Water in the Cathode Compartment of a Biofuel Cell," *J. Electroanal. Chem.*, **464**, 110 (1999).
- Park, S., R. Craciun, J. M. Vohs, and R. J. Gorte, "Direct Oxidation of Hydrocarbons in a Solid Oxide Fuel Cell I. Methane Oxidation," *J. Electrochem. Soc.*, **146**, 3603 (1999).
- St-Pierre, J., D. P. Wilkinson, S. Knights, and M. L. Bos, "Relationships between Water Management, Contamination and Lifetime Degradation in PEFC," *J. New Mat. Electrochem. Systems*, **3**, 99 (2000a).
- St-Pierre, J., S. A. Campbell, M. K. Watson, M. P. Sexsmith, M. Derflinger, and G. Hornburg, "Antifreeze Cooling Subsystem," PCT application WO 00/17951 (Mar. 30, 2000b).
- Stumper, J., S. A. Campbell, D. P. Wilkinson, M. C. Johnson, and M. Davis, "In-situ Methods for the Determination of Current Distributions in PEM Fuel Cells," *Electrochim. Acta*, **43**, 3773 (1998).
- Weidner, J. W., and M. Doyle, "Report on the Electrolytic Industries for the Year 1999," *J. Electrochem. Soc.*, **147**, 3953 (2000).
- Wilkinson, D. P., H. H. Voss, J. Dudley, G. J. Lamont, and V. Basura, "Electrochemical Fuel Cell Assembly with Integral Selective Oxidizer," U.S. Patent No. 5,482,680 (Jan. 9, 1996).
- Wilkinson, D. P., B. H. Chiem, J. A. Roberts, J. St-Pierre, J. Stumper, and J. Zimmermann, "Method and Apparatus for Controlling the Temperature within an Electrochemical Fuel Cell," PCT application WO 00/33407 (Nov. 29, 1999).
- Wilkinson, D. P., and J. St-Pierre, "In-plane Gradients in Fuel Cell Structure and Conditions for Higher Performance," *J. Power Sources*, in press (2001).
- Winsel, A., "Method of Removing Water of Reaction during Fuel Cell Operation," U.S. Patent No. 3,553,026 (Jan. 5, 1971).
- Wozniczka, B., N. J. Fletcher, and P. R. Gibb, "Electrochemical Fuel Cell Stack with Compression Bands," U.S. Patent No. 5,789,091 (Aug. 4, 1998).

