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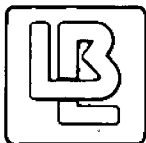
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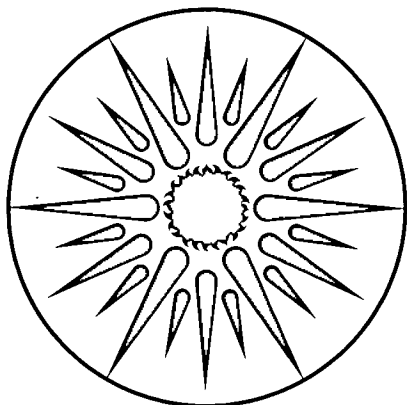
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Fuel Cell Technology in the United States

K. Kinoshita

April 1989

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FUEL CELL TECHNOLOGY IN THE UNITED STATES

by

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FUEL CELL TECHNOLOGY IN THE UNITED STATES

Purpose

Fuel cells provide a new and exciting option for the efficient conversion of fossil fuels to electricity. Development of fuel-cell technology has been underway in the United States (US) for over 20 years, with the initial focus on space applications (*i.e.*, Gemini, Apollo). More recently, commercialization of fuel cells for utility power generation and transportation applications has received considerable attention in the US. The purpose of this paper is to summarize the status of fuel-cell technology in the US, where development of five major fuel cell systems are underway.

Introduction to Fuel Cells

A recent review by Kinoshita et al. (1) provides a summary of fuel-cell technology. In this section, a brief overview is provided as an introduction for readers who may not be familiar with this technology. At last count, there were over 40 books that described various aspects of fuel-cell technology, and these are listed in Reference 1.

Fuel cells are electrochemical devices that convert the chemical energy of a reaction directly into electrical energy. In a typical fuel cell, gaseous fuels are fed continuously to the anode (negative electrode) compartment and an oxidant (*i.e.*, oxygen from air) is fed continuously to the cathode (positive electrode) compartment; the electrochemical reactions take place at the electrodes to produce an electric current. The fuel cell is an energy conversion device which theoretically has the capability of producing electrical energy for as long as the fuel and oxidant are fed to the electrodes. In reality, degradation or malfunction of components limits the practical operating life of fuel cells.

A variety of fuel cells have been developed, and they are usually classified according to the type of electrolyte used in the cells; these include i) polymer electrolyte fuel cell (PEFC), ii) alkaline fuel cell (AFC), iii) phosphoric acid fuel cell (PAFC), iv) molten carbonate fuel cell (MCFC), and v) solid oxide fuel cell (SOFC). These fuel cells are listed in the approximate order of increasing operating

temperature, ranging from -80°C for PEFC, $\sim 100\text{-}200^{\circ}\text{C}$ for AFC, $\sim 200^{\circ}\text{C}$ for PAFC, $\sim 650^{\circ}\text{C}$ for MCFC and $\sim 1000^{\circ}\text{C}$ for SOFC.

The basic elements of a fuel cell are the electrolyte phase, and contacting porous anode and cathode on either side. Figure 1 shows simplified schematic representations of a single cell and a fuel cell integrated with the ancillary components that constitutes a fuel-cell plant. In the fuel cell, the fuel and oxidant gases flow past the backside of the anode and cathode, respectively, and generate electrical energy by the electrochemical oxidation of fuel, usually hydrogen, and the electrochemical reduction of oxygen. A three-phase region (porous electrode/electrolyte/reactant interface) provides the extensive reaction area that is required to achieve practical current densities in fuel cells. The nature of this three-phase region plays a critical role in the electrochemical performance of a fuel cell, particularly in those fuel cells with liquid electrolytes (*i.e.*, AFC, PAFC, MCFC). In such fuel cells, the reactant gases diffuse through a thin electrolyte film that wets portions of the porous electrode and react electrochemically on the electrode surface. If the porous electrode contains an excessive amount of electrolyte, the electrode may “flood” and restrict the transport of gaseous species in the electrolyte phase. The consequence is a reduction in the electrochemical performance of the porous electrode. Thus a delicate balance must be maintained among the electrode, electrolyte and gaseous phases in the porous electrode structure.

The typical cell components used in various fuel-cell systems are listed in Table 1. Fuel cells that operate at low temperatures (*i.e.*, $\leq 200^{\circ}\text{C}$) utilize components made of metals, alloys, carbonaceous materials and polymers. However, fuel cells operating at higher temperatures require components that contain ceramic metal oxides, both electrically conducting (ionic, electronic) and insulators, depending on the intended application. As the operating temperature of fuel cells is raised, a severe limitation is imposed on the options available with regard to materials selection for the various components. The physicochemical and thermomechanical properties of materials used in the cell components (*i.e.*, electrodes, electrolyte, separator, current collector, etc.) determine the practical operating temperature and useful life of a fuel cell. The operating temperature of fuel cells with

aqueous electrolytes (and polymer electrolytes) is limited to about 200°C or lower because of the high water vapor pressure and/or rapid degradation of cell components at higher temperatures.

A fuel-cell stack usually consists of a number of individual cells that are electrically connected in series and manifolded for the fuel and oxidant gases and for thermal management of excess heat. The stack is usually integrated to two major subsystems to form a fuel-cell plant (see schematic representation in Figure 1): i) a power conditioner to convert dc power to ac power and ii) a fuel processor unit to convert commercially available fuels into a fuel that is effectively utilized in the cell. The integration of the fuel processor, fuel cell, power conditioner and other ancillaries (*e.g.*, turbocompressors and blowers, thermal management subsystem, bottoming cycle subsystem to generate additional electricity and/or heat recovery for other use such as for heating water, space heating, low pressure steam, etc.) must be optimized to achieve the maximum efficiency from the system.

Two structural designs are commonly used in fuel-cell stacks: i) flat-plate and ii) tubular. In the flat-plate design (see Figure 2) the cell components are thin, flat structures that are arranged in a parallel sequence to form a cell. The individual cells are separated by a bipolar separator which has two major functions: i) current collector—provides the electrical series connection between cells, and ii) gas barrier—separates the fuel and oxidant streams in adjacent cells. The bipolar plate must be both an electrical conductor and impervious to gases. This so-called bipolar arrangement minimizes the ohmic resistance in the fuel-cell stack because the current-conduction path is short through the electrolyte and other cell components. The tubular configuration, which is discussed in a later section, requires a more complex electrical connection to form a fuel-cell stack, and the ohmic resistance in the cell tends to be a problem. In the representation shown in Figure 2, the fuel and oxidant streams flow perpendicular to each other (*i.e.*, “crossflow”) and the current flows perpendicular to the gas flow. Other arrangements of gas flow and current flow are also used in fuel-cell stack designs.

Low-temperature fuel cells (PEFC, AFC, PAFC) require electrocatalysts to achieve practical reaction rates at the anode and cathode, and hydrogen is the only acceptable fuel for large-scale fuel-

cell power plants because CO inhibits the anode reaction. With high-temperature fuel cells (MCFC, SOFC), the requirements for electrocatalysis are relaxed, and the number of potential fuels is increased. Besides H₂, CO and CH₄ are potential fuels because they undergo chemical reaction with H₂O to produce H₂ which is electrochemically oxidized. In MCFCs, CO and CH₄ are sources of H₂ from water-gas shift and steam-reforming reactions, respectively. The direct oxidation of CO and CH₄ in high-temperature SOFCs is feasible, but they are still not as easily oxidized as H₂.

The typical half-cell reactions that occur in practical fuel cells are summarized in Table 2. The overall electrochemical reactions corresponding to the individual electrode reactions are:



with H₂ and O₂ reacting in fuel cells such as PEFC, AFC, PAFC and SOFC to produce H₂O. When a carbon-containing fuel is involved in the anode reaction, CO₂ is also produced. In the case of MCFCs, CO₂ is required in the cathode reaction to maintain an invariant carbonate concentration in the electrolyte. The overall cell reaction in MCFCs is given by



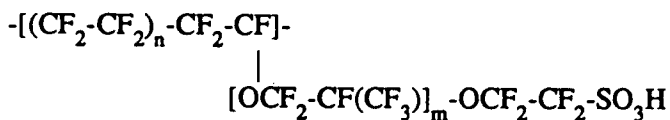
where a and c refer to the anode and cathode compartments, respectively. Besides the reaction of H₂ and O₂ to produce H₂O, CO₂ is transferred from the cathode compartment to the anode compartment, with 1 mole CO₂ transferred along with 2 Faradays of charge. In a practical MCFC, the CO₂ generated at the anode is recycled to the cathode where it is consumed.

Polymer Electrolyte Fuel Cell

The use of cation-exchange membrane polymers as the electrolyte in fuel cells was originally conceived by Grubb (2) in the 1950s. The first PEFCs were demonstrated in the early stages of the US Space Program (Gemini) in the 1960s. More recently, the primary emphasis of this technology in the US is to develop PEFCs for terrestrial applications in transportation.

The early membranes tested in PEFCs include hydrocarbon-type polymers such as cross-linked polystyrene-divinylbenzene sulfonic acids and sulfonated phenolformaldehyde. The Gemini Space

Program [fuel cell module: 31.7-cm diameter, 63.5-cm high, 30 kg, 1 kW at 23.3-26.5 V; cell operated at 37 mA/cm² at 0.78 V on pure H₂ and O₂ at 138-207 kPa absolute (20-30 psia) and ~35°C] in the 1960s employed membranes of polystyrene-divinylbenzene sulfonic acid crosslinked to an inert fluorocarbon film. It was observed that the life of PEFCs was limited by oxidative degradation of the polymer. Hydrocarbon-type polymers are unstable because of cleavage of the C-H bonds, particularly the α-H where the functional group is attached. When these polystyrenes were replaced with fluorine-substituted polystyrenes (*e.g.*, polytrifluorostyrene sulfonic acid), the life of PEFCs was extended by four to five times. However, the operating temperature of PEFCs with fluorinated polystyrenes was limited to less than 75°C. Research started in the early 1960s led to the development of Nafion (registered trademark of E.I. DuPont de Nemours), which are perfluorosulfonic acid membranes that are electrochemically stable in PEFCs at temperatures up to about 100°C. These polymer consists of the following ionomer units,



where $n = 6-10$ and $m \geq 1$. Nafion and its derivatives all have two features in common: i) polymer chains consist mainly of a PTFE backbone, which statistically form segments several units in length, and ii) perfluorinated vinyl polyether, a few ether links long, which joins these segments to form a flexible branch pendant to the main perfluoro-chain and carries a terminal acidic group to provide the cation-exchange capacity. These perfluorinated ionomer membranes with sulfonic acid groups meet all the required characteristics of ion-exchange membranes for use in fuel cells, as well as for use in H₂O and alkali hydroxide electrolysis cells. Nafion was first used in fuel cells in 1966, and it is still the most widely tested ion-exchange membrane in PEFCs.

A new series of perfluorinated ionomers has recently become available from Dow Chemical Company which provides an attractive alternative to Nafion in PEFCs (3,4). This new polymer has a PTFE-like backbone similar to that of Nafion, but the pendant side chain containing the sulfonic acid

group is shorter. Instead of the long side chain of Nafion shown above, the Dow polymer has a side chain consisting of $-\text{OCF}_2-\text{CF}_2-\text{SO}_3\text{H}$. This polymer possesses ion-exchange properties similar to that of Nafion, and it is also available with higher acid strength and lower equivalent weights (*i.e.*, 600-950). Even at these low equivalent weights the Dow membrane has good mechanical strength and does not hydrate excessively, whereas Nafion of comparable equivalent weight would form a highly gelled polymer having poor or no mechanical integrity. The Dow polymer has a higher glass transition temperature (165°C versus 110°C for Nafion), thus PEFCs containing this material should be capable of operating at higher temperatures (>100°C).

The early PEFCs containing Nafion membranes utilized about 4 mg/cm² of Pt black in both cathodes and anodes. Such electrode/membrane combinations, with the appropriate current collectors and supporting structure in PEFCs were capable of operating at pressures up to 20 MPa, differential pressures up to 3.4 MPa, and current densities of 2000 mA/cm² (5). In space applications where cost is not of primary concern, high Pt loadings in the electrode are tolerable. However, for terrestrial applications such as in electric vehicles, high Pt loading would make the cost prohibitive.

Several recent developments have improved the prospects for PEFCs in terrestrial applications. These developments are i) the reduction of noble-metal electrocatalyst in the electrodes and ii) the development of an alternative ion-exchange membranes for PEFCs. Electrodes with 0.35 mg Pt/cm², and impregnated with a proton-conducting polymer, have exhibited comparable activity for O₂ reduction (600 mA/cm² at 570 mV, IR-free versus RHE, 0.1 MPa) as earlier electrodes with much higher Pt loadings. The synthesis of perfluorinated sulfonic acid membranes by Dow Chemical Company, which appears to possess superior water-management properties to that of Nafion, is expected to improve the prospects for the successful implementation of PEFCs in electric vehicle applications. However, it should be noted that even at low Pt loadings, about \$1000 (US) of Pt (assuming Pt costs \$14/g) would still be required in PEFCs of the size needed for automobile propulsion (6).

Alkaline Fuel Cell

Because alkaline electrolytes (*i.e.*, NaOH, KOH) do not reject CO₂, AFCs are currently considered in the US for specialized applications where pure H₂ and O₂ are utilized. However, efforts are underway in Europe to utilize AFCs in fleet applications of electric vehicles (*i.e.*, city buses, garbage trucks). In these applications, the alkaline electrolyte is circulated to minimize the accumulation of carbonates and to facilitate thermal management.

The AFC used in the US Apollo Space Program was based, in large part, on the technology originally developed by Bacon (7) in the 1930s. His fuel cells operated at 200-240°C with 45% KOH, and the pressure was maintained at 4-5.5 MPa to prevent the electrolyte from boiling. The electrodes used in the US Apollo Program were nickel-based dual-porosity structures (two-layer structure with porous Ni of 16- μ m maximum pore diameter on the electrolyte side and 30- μ m pore diameter on the gas side), and the cathode consisted of a porous structure of lithiated NiO. The three-phase boundary in the porous electrodes was maintained by a differential gas pressure across the electrode since a wetproofing agent was not available. These AFCs (fuel-cell module: 57-cm diameter, 112-cm high, ~110 kg, 1.42 kW at 27-31 V, 0.6-kW average power) utilized pure H₂ and O₂ and highly concentrated electrolyte (85% KOH) to permit cell operation at lower pressure (~414 kPa reactant gas pressure) without electrolyte boiling. In this concentrated electrolyte, the cell performance was not as high as in less concentrated electrolyte, consequently the operating temperature was increased to 260°C. The typical performance of this AFC was 0.85 V at 150 mA/cm², which compared favorably to the performance of the Bacon cell operating at about 10 times higher pressure.

The electrodes utilized in the US Space Shuttle Orbiter contain high loadings of noble metals [anode: 10 mg/cm² (80% Pt/20% Pd) on a Ag-plated Ni screen, cathode: 20 mg/cm² (10% Pt/90% Au) on a Ag-plated Ni screen] that are bonded with PTFE to achieve high performance at lower temperature. The AFC used in the Space Shuttle Orbiter (fuel-cell module: 35-cm high, 38-cm wide, 101-cm long, 91 kg, 12 kW at 27.5 V, 7-kW average power) operates in the same pressure range as

those in the Apollo Program but at a lower temperature (80 to 90°C) and a higher current density (470 mA/cm² at 0.86 V).

Fuel cells, and AFCs in particular, are being considered in power systems for US military applications, *i.e.*, the Strategic Defense Initiative (SDI). Projections indicate that AFCs and SOFCs can meet the SDI performance goals of specific power of 3.2 kW/kg and specific energy of 444 Wh/kg (8). To reach these goals with AFCs will require a lower cell weight, which is achieved by eliminating separate liquid-cooling channels, utilizing lightweight components, and developing thinner cell components. The goal for total cell thickness is 0.57 mm. In addition, operation of the fuel cell at a temperature of 150°C and a pressure of 1.37 MPa (200 psia) on H₂ and O₂ is expected to provide the performance to meet the goals.

Phosphoric Acid Fuel Cell

The development of cell components for PAFCs has undergone evolutionary changes from the mid 1960s to the present as this fuel-cell technology approaches commercialization by US and Japanese developers. In the mid 1960s, the conventional porous electrodes were PTFE-bonded Pt black, and the loadings were about 9 mg/cm² Pt. One of the major breakthroughs in PAFC technology that occurred in the late 1960s was the development of carbon blacks and graphites for cell components. These materials were sufficiently stable to replace the more expensive gold-plated tantalum cell hardware. Because of the availability of low-cost carbon blacks (*e.g.*, furnace black, acetylene black), dramatic changes in the development of porous electrodes, with Pt supported on carbon black replacing Pt black in porous PTFE-bonded electrode structures as the electrocatalyst. Currently, the Pt loadings are, about 0.25 mg/cm² Pt in the anode (Pt may be alloyed with other metals) and about 0.50 mg/cm² Pt in the cathode. Assuming a cell voltage of 700 mV at 200 mA/cm² and the current Pt loadings at the anode and cathode, ~5.36 g Pt/kW of power generated is required.

Transition metal (*e.g.*, iron, cobalt) organic macrocycles from the families of tetramethoxyphenylporphyrins (TMPP), phthalocyanines (PC), tetraazaannulenes (TAA) and tetraphenyl-

porphyrins (TPP) have been evaluated as O₂-reduction electrocatalysts in PAFCs. One major problem with these organic macrocycles is their limited chemical stability in hot concentrated phosphoric acid. However, after heat-treatment of the organic macrocycle (*i.e.*, CoTAA, CoPC, CoTMPP, FePC, FeTMPP) on carbon at about 500 to 800°C, the pyrolyzed residue exhibits electrocatalytic activity, which in some instances, is comparable to that of Pt, and also has promising stability at lower temperatures (*i.e.*, ~100°C) in acid electrolytes (9). Another approach that has been successful for enhancing the electrocatalysis of O₂ reduction is to alloy Pt with transition metals such as Ti (10), Cr (11,12), V (13,14), Zr (13) and Ta (13).

By the mid 1980s, reasonable long-term endurance was demonstrated for PAFC stacks in the US. Thirty-cell stacks (0.34 m² electrode area) have operated at 205°C and 0.83 MPa for >16,000 h at 216 mA/cm². A 20-cell stack with larger electrodes (0.98 m² electrode area) has operated for over 1000 h at at 205°C and 0.83 MPa with a decay rate of only 3-4 mV/1000 h; both the performance and decay rate are within the accepted goal (*i.e.*, performance at 1000 h of 0.76 V at 216 mA/cm², end-of-life performance of 0.73, decay rate of <6 mV/1000 h) for PAFCs for utility applications.

Practical pressure-temperature combinations for operating PAFCs are: 0.3 MPa at 190°C, 0.6 MPa at 210°C and 0.8 MPa at 220°C (15). The initial cell voltage of a PAFC operating at 190°C and 0.3-MPa pressure is about 38 mV higher than a PAFC at atmospheric pressure and 190°C. At 220°C and 0.8-MPa pressure, an even greater improvement in performance is obtained, particularly with a Pt-V alloy rather than Pt in the cathode.

International Fuel Cells Corp. (IFC) and Westinghouse Electric Corporation (WEC) are the two US industrial developers of PAFCs for utility power generation. One major difference in their PAFC technologies is the method of thermal management used in the cell stacks. IFC has elected to use a liquid (two-phase water) to remove heat that is generated during cell operation, whereas WEC is using gas (air) cooling. In either case, the coolant is pumped through cooling channels located (usually between every fifth cell) in the cell stack. Liquid cooling requires complex manifolding and con-

nections, but better heat removal is achieved than with air cooling. However, the advantage of air cooling is its simplicity, reliability and relatively low cost.

Other differences in the PAFC stack design of IFC and WEC are the gas distribution channels and the configuration of the bipolar plate separator. The "ribbed substrate" configuration was developed by IFC to reduce cost and to provide a storage reservoir for additional acid. In this design concept, the traditional bipolar separator (see Figure 2) with gas channels on both sides is replaced by several components; a thin impervious plate serves to separate the reactant gases in adjacent cells in the stack, and separate porous plates with ribbed channels on only one face are used for directing gas flow. In a cell stack the impervious plate is located between the two porous plates, and in contact with the ribbed channels. The electrocatalysts are placed on the smooth face of the porous plates, and electrolyte is stored in the ribs of the porous plate. By machining ribbed channels on only one side of the plate, a significant cost savings is achieved. Furthermore, the ribbed substrate permits about a fivefold increase in effective electrolyte volume in the stack compared to the conventional bipolar cells. This added electrolyte replaces the acid lost from the cell during operation and is sufficient to achieve cell life of more than 40000 h at 205-210°C and 0.82 MPa pressure. The gas flow channels in the WEC design do not pass straight through the cell (see Figure 2). Instead the channels form a "zee pattern. In this Z-plate design, the inlet for the oxidant gas and exit for the fuel gas are side by side on one end of the narrow side of the rectangular bipolar structure. The exit for the oxidant gas and the inlet for the fuel gas are side by side, but on the opposite half of the other narrow side (for a counterflow arrangement). This pattern is achieved by having the gas channels make a right-angle turn so they are directed to the side of the bipolar structure, followed by a left-angle turn to direct the gas channel to the opposite narrow side. The other reactant gas would have channels that make opposite turns before reaching the other side of the bipolar structure. Cooling air passes through the bipolar structure channels that traverse the short dimension in the rectangular stack structure.

Molten Carbonate Fuel Cell

One difference between low-temperature fuel cells (AFC, PAFC) and MCFCs is the method used to retain electrolyte in the respective cells. In AFCs and PAFCs, PTFE serves as a binder and wet-proofing agent to maintain the integrity of the electrode structure and to establish a stable electrolyte interface in the porous electrodes. There are no materials available for use in MCFCs which can serve a similar function to PTFE in low-temperature fuel cells. Thus, the approach used to maintain a stable electrolyte interface in MCFC porous electrodes is to rely on a balance in capillary pressures of the molten electrolyte in the electrode and electrolyte matrix (16,17,18). By properly matching the pore diameters in the electrodes with the electrolyte matrix, which contains the smallest pores, the electrolyte distributes so that the electrolyte matrix remains completely filled with molten carbonate, while the porous anode and cathode are partially filled, depending on their pore-size distributions.

In the mid 1960s, the typical electrode materials used in MCFC tests were Fe, Cu, Ni, stainless steels and precious metals (*e.g.*, Pt, Au, Ag), but the majority of these tests were conducted in small laboratory cells. The technology soon evolved to the use of Ni-based alloys and oxides, and since the mid 1970s, the materials for the electrodes and electrolyte structure (molten carbonate/LiAlO₂) have remained essentially unchanged. A major development in the US over the last 10 years has been the evolution in the technology for fabrication of electrolyte structures.

Currently, porous lithiated NiO is the conventional cathode material in MCFCs. The NiO is usually formed *in situ* in MCFCs by oxidation of porous Ni, which results in ~30% volume expansion. This procedure results in poor control to the physical properties (*i.e.*, porosity, pore size distribution, mean pore size) of the oxidized nickel structure; consequently matching the electrode pore structure to the pore distribution in the electrolyte matrix is difficult. Furthermore, the NiO structure formed *in situ* possesses poor mechanical integrity, and therefore it is susceptible to further degradation in MCFCs. A major problem that was discovered with NiO cathodes in MCFCs is dissolution of Ni. Despite its low solubility (~10 ppm) in molten carbonates, Ni ions diffuse in the electrolyte

towards the anode and metallic Ni can precipitate in regions where a H₂ reducing environment is encountered. The net result is the formation of dendritic Ni which can produce a short-circuited fuel cell. The rate of Ni dissolution is enhanced at higher partial pressures of CO₂ in the cathode gas. However, the goal of 40,000 h for the lifetime of MCFCs appears achievable with cell operation at atmospheric pressure and with CO₂ partial pressures of less than -0.015 MPa, and if the cell is rarely at open-circuit potential. Because of the physical and chemical limitations of NiO, research is underway to develop alternative cathode materials, and two metal oxides, Cu- and Co-doped LiFeO₂ and Mg-doped Li₂MnO₃, have emerged as leading candidates (19,20).

Nickel-based alloys (*e.g.*, Ni-10% Cr) are the conventional anode materials. A major problem with these materials is their structural stability. The compressive forces in MCFCs results in mechanical deformation of the porous Ni-based anodes which leads to severe performance decay as the electrolyte is redistributed in the MCFC stack. Current approaches to overcome this problem include the addition of metal oxides (*i.e.*, oxide-dispersion-strengthened anodes) to the porous Ni-based anode which improves its mechanical properties.

The MCFC is particularly sensitive to impurities in the fuel and oxidant gas streams. It is now well-established that sulfur compounds in the low-ppm concentrations in fuel gases are detrimental because they react with Ni and the molten carbonate electrolyte. Other contaminants can also have a damaging effect on MCFC performance and life; these are summarized by Kinoshita et al. (1).

In the 20 years from the mid 1960s to the mid 1980s, the performance of single cells has improved from about 10 mW/cm² to >150 mW/cm². Since the early 1980s both the performance and endurance of MCFC stacks have shown dramatic improvements, but they have not reached the endurance achieved with PAFCs. Single cells have been tested for over 13,000 h and 20-cell stacks (0.72 m² electrode area, 20 kW) have operated successfully for over 1700 h.

Various options in cell and stack design are being pursued by MCFC developers in the US. International Fuel Cells and Energy Research Corporation have adopted the traditional concept of

cells arranged to form stacks, as depicted schematically in Figure 1. The reactant gases are introduced in parallel to the cells through common manifolds on the side of the stack, with the fuel and oxidant gases flowing in a cross-flow pattern (*i.e.*, at right angles to each other) through the cell. In MCFCs developed by IFC, fuel processing is performed external to the stack, while ERC has pursued internal reforming of the fuel gas (IRMCFC). With internal reforming, the need for a separate, external fuel processor and its ancillary equipment is eliminated. In other words, the fuel-cell system is designed so that fuel processing occurs on reforming catalysts near the electrochemical sites in the MCFC stack. Two key issues with IRMCFCs are to integrate the reforming and electrochemical reactions and to maintain the proper thermal balance in the cell stack. A preliminary analysis suggests that a 1.8-MW IRMCFC using natural gas should be capable of achieving an efficiency of 52.9%.

MC-Power is developing another design concept for MCFCs. Their approach differs from those of IFC and ERC in that the fuel and reactant gases are internally manifolded. The design of the single-piece separator plate/current collector that is used for internal manifolding is analogous to plate-type heat exchangers. One advantage of internal manifolding is that it eliminates carbonate migration present with gaskets used for external manifolding. It is also claimed by the developers that internal manifolding, because of its unique geometric pattern, permits more efficient cell-to-cell gas distribution and heat exchange.

Solid Oxide Fuel Cell

The SOFC technology has emerged in the 1980s from scale up of single cells to prototype multi-cell arrays. The SOFC, besides its extremely high temperature (~1000°C) of operation, differs from other conventional fuel cells (AFC, PAFC, MCFC) in that a liquid electrolyte is not required. Instead, the electrolyte is a solid, and thus electrolyte-management problems that are common to the other conventional fuel cells are not a concern for SOFCs. In addition, SOFCs do not require CO₂ recycle from the anode to the cathode, such as in MCFCs. Active programs are underway in the US to develop SOFCs of tubular geometry, as well as bipolar stack designs.

In the early 1960s, experimental SOFCs with planar geometry were evaluated, but this geometry presented a problem for building cell stacks because of difficulties with obtaining adequate gas seals. A tubular configuration (*i.e.*, cylindrical design) was adopted for SOFCs which appeared to alleviate the problems with gas seals, and this design is currently used in the development program at Westinghouse Electric Company. An early tubular design consisted of overlapping components (*i.e.*, electrodes, electrolyte, cell interconnection) in thin layers (10-50 μm), which were deposited on a porous support tube of calcia-stabilized zirconia (21,22,23). In this tubular design, individual fuel cells are arranged in bands along the support tube and are connected in series by a ceramic interconnect material. Another variation of an early tubular design is referred to as a "bell and spigot" configuration, which consists of short, cylindrical electrolyte segments shaped so that they can be fitted one into the other and connected into a long tube by bell-and-spigot joints (24,25). A schematic representation of the cross section of the present tubular design for a SOFC is presented in Figure 4. In this design the cathode, cell interconnection, electrolyte and anode are sequentially deposited on a closed-end porous tubular support material which provides a mechanically strong structure for the thin cell components. The oxidant gas is introduced via a central Al_2O_3 tube, and the fuel gas is supplied to the exterior of the closed-end tube. The present tubular design uses a porous support tube of about 30-cm length and 1.27-cm diameter, with an active area of about 110 cm^2 . These cells produce about 18 W each, thus about 55 cells are required to generate 1 kW.

A bipolar structure, which is the common configuration for cell stacks in PAFCs and MCFCs, permits a simple series electrical connection between cells. Perpendicular current collection in a cell stack with a bipolar design should have a lower ohmic polarization than the tubular configuration, and overall stack performance should be improved. However, gas leaks in SOFCs of bipolar configuration with compressive seals are difficult to prevent, and thermal stresses at extended interfaces between dissimilar materials must be accommodated to prevent mechanical degradation of cell components. Variations in planar electrodes and solid electrolyte structures are being developed by Garrett/AiResearch and Combustion Engineering, International Fuel Cells Inc. and Ceramatec, and a

small effort is underway at Ztek Corporation.

The so-called monolith structure is a more complex design of bipolar configuration for SOFCs. The design was conceived at Argonne National Laboratory, and industrial development is underway at Garrett/AiResearch and Combustion Engineering. A schematic representation of a cross section of the monolith structure proposed for SOFCs is presented in Figure 5. The structure resembles the corrugated assembly used in cardboard boxes. The small channels are formed from thin (25 to 100 μm) layers of the active cell components, and these channels serve for gas passage of the fuel and oxidant streams.

Both geometric designs for SOFCs share the same types of materials in the cell components (see Table 1). The anodes consist of nickel cermet (metallic Ni and Y_2O_3 -stabilized ZrO_2 , which serves to inhibit sintering of the metal particles), and the cathodes are Sr-doped lanthanum manganite ($\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, $x = 0.010\text{--}0.15$), which is a p-type conductor. Zirconia-based electrolytes are suitable for SOFCs because they exhibit pure anionic conductivity over a wide range of oxygen partial pressures (0.1 to 10^{-21} MPa). The electrolyte must have a transport number for O^- of close to unity as possible, and a transport number for electronic conduction of close to zero as possible. The other cell components should permit electronic conduction, and interdiffusion of ionic species in these components at 1000°C should not have a major effect on their electronic conductivity. The cell interconnection material must be impervious to fuel and oxidant gases, and thus it must be chemically stable under oxygen partial pressures of about ~ 0.1 to 10^{-19} MPa at 1000°C .

The thermodynamic efficiency ($\Delta G/\Delta H$) of SOFCs is lower than that of MCFCs and PAFCs which utilize H_2 and O_2 because of the lower ΔG at higher temperatures; ΔG decreases from 54.617 kcal/mole at 27°C to 43.3 kcal/mole at 927°C , whereas ΔH is nearly constant over this temperature range. However, the higher operating temperature of SOFCs is beneficial in reducing polarization losses. The polarization in SOFCs is dominated by ohmic losses (IR) in the cell components, with the major contribution in a tubular cell originating from the cathode (*i.e.*, 65% of the total). The

cathode IR dominates the total ohmic loss despite the higher specific resistivities of the electrolyte and cell interconnection because of the short conduction path through these components and the long current path (*i.e.*, 1.1 cm) in the plane of the cathode. The current path in the anode is about 0.8 cm and its resistivity is about an order of magnitude lower than that of the cathode, so for typical thickness of these electrodes the IR loss in the cathode is about 2.5 times greater than in the anode. Consequently the IR drop by current collection around the cell perimeter puts an upper limit on tube diameter for acceptable performance. Current collection and IR loss in the cathode is not expected to limit the performance in SOFCs with planar geometry because of the short conduction paths (*i.e.*, perpendicular current collection). Instead, the major challenge with the bipolar configuration using the planar geometry is to fabricate cell arrays with acceptable intercell conductivity and thermomechanical properties.

Small SOFCs (9-cm² active area) with the monolith geometry have been fabricated and tested at 1000°C (26). Current densities as high as 2.2 A/cm² have been achieved on hydrogen and air, and an array consisting of two cells in series has operated for more than 650 h at 50 mA/cm² and -0.6 V. These cells were also operated with hydrocarbon fuels (*e.g.*, natural gas, CH₄, C₃H₈, C₈H₁₈, C₂H₅OH, natural gas) that were mixed with H₂O, and the performance was similar to that obtained with hydrogen. Furthermore, the presence of about 5 ppm sulfur in the natural gas had no detrimental effect on cell performance. The cell performance obtained to date indicates that the ohmic resistance is the limiting factor. In particular the interfacial resistance at the fuel electrode/electrolyte interface needs to be reduced further by optimizing the distribution of Ni. The successful scale up of the small monolith arrays into larger size stacks (*i.e.*, larger area and greater number of cells in series connection) that have a long life has not been demonstrated.

Technology Status

Fuel-cell technology in the US has evolved from small laboratory cells to fuel-cell systems that are on the verge of commercialization. From the mid 1960s to the mid 1980s an order-of-magnitude

increase in the size of cell stacks and an order-of-magnitude increase in demonstrated stack durability have occurred. Over the same time period, approximately one order-of-magnitude improvement in fuel-cell performance, as measured by the practical power density of fuel-cell stacks, has also occurred.

Figure 5 provides a comparison of cell performance levels (expressed as current-voltage curves) which are representative of current fuel-cell technology for different systems. In this example, the AFC performance is based on pure H_2 which is dead-ended in the cell (*i.e.*, 100% fuel utilization), while the other fuel cells are operated at 85% fuel utilization. The performance of the PAFC is typical of the single cell performance in PC-23, which is the designation by IFC for stacks that are available for their 11-MW PAFC plant. The lower performance obtained with MCFCs and SOFCs (projected with internal reforming of CH_4) reflects the thermodynamic penalty incurred by high-temperature fuel cells. However, the efficiency of the high-temperature fuel cells are higher than that of PAFCs. The efficiency (E_{fc}) is equal to $V_c U / HHV$, where V_c is the cell voltage, U is the fuel utilization, and HHV is the higher heating value of the fuel. For H_2 and CH_4 , the voltage equivalents for HHV are 1.48 and 1.14 V, respectively. For PAFCs operating at $V_c = 0.7$ V and $U = 85\%$, $E_{fc} = 40\%$ $[(0.7 \times 0.85)/1.48]$. The corresponding efficiency of a high-temperature fuel cell operating at the same V_c and U is 52% $[(0.7 \times 0.85)/1.14]$, with internal reforming of CH_4 . An AFC operating at a higher cell voltage, say $V_c = 0.8$ V, and at $U = 100\%$ with pure H_2 provides an opportunity to obtain $E_{fc} = 54\%$. Unfortunately, reforming fossil fuels to obtain pure H_2 incurs an efficiency penalty that reduces the overall AFC plant efficiency to lower than that of the high-temperature fuel cells.

Table 4 provides a brief summary of the major US industrial participants involved in fuel-cell technology. Only one company, IFC, has an active program in each of the five fuel-cell technologies. Their fuel-cell programs are directed at space, military, utility and transportation applications. A recent entry into fuel-cell technology is General Motors Corporation, which is interested in developing PEFCs for transportation applications. Two major US chemical companies, Dow Chemical

Company and Du Pont, Inc. (E.I. Du Pont de Nemours & Company, Inc.), have established a presence in PEFC technology with their development of ion-exchange membranes. Several smaller companies are developing their PEFC technologies for specialty markets. Westinghouse Electric Corporation is pursuing the development of PAFC for utility applications. Their PAFC technology is partially based on R&D which was conducted at Energy Research Corporation. Westinghouse has also been actively involved in developing tubular SOFCs. More recently, Combustion Engineering and Garrett/AiResearch (Division of Allied-Signal Aerospace Company) have entered into a program to develop monolith SOFCs, which were originally conceived at Argonne National Laboratory. Three industrial companies, with different design concepts are involved in developing MCFCs for utility power applications. Both IFC and ERC have extensive experience in MCFC technology, dating back to the early 1970s. M-C Power Corp. is a subsidiary of the Institute of Gas Technology, which has a long-standing program in MCFC technology, dating back to the 1960s. They also have a collaborative agreement with Combustion Engineering, Inc., as well as with European and Japanese companies.

Only a limited number of endurance tests of fuel-cell stacks have been conducted by US developers, and some of the major ones are listed in Table 5. The earliest demonstrations were the AFCs in the US Space Program, which were of reasonable short duration compared to projections for fuel cells in terrestrial applications. The design life of the AFCs in the Space Shuttle Orbiter is about 2000 h, about 5 times longer than that for the Apollo Program. IFC has tested various sizes of PAFCs at many different sites. Their largest demonstration (4.5-MW AC) took place in Japan (Tokyo Electric Power, Inc., Goi), and operational tests started on 7 April 1983, with rated power achieved in February 1984 on natural gas. The fuel-cell tests lasted about two years (ended December 1985), and reached a performance of 0.65 V/cell at 250 mA/cm² (190°C, 0.34 MPa), which corresponds to an efficiency of 37% (HHV). Tests of small MCFC stacks (*e.g.*, 7200-cm² electrode area, 20 cells, 20-kW stack by IFC; 3700-cm² electrode area, 11 cells, 5 kW by ERC; 1000-cm² electrode area, 10 cells, 1 kW by M-C Power) have been conducted by MCFC developers, but experience here is

restricted to in-house tests of only about 2000 h. With regard to SOFC tests, a 3-kW system (144 tubular cells, 36-cm active length) consisting of tubular cells fabricated by Westinghouse was tested in Japan. By the middle of 1988, one of the 3-kW systems reached 5000 h of operation.

The characteristics of various fuel-cell systems have been evaluated and compared in numerous studies. These studies indicate that the cost of electricity (COE) will be lower with MCFC and SOFC than with PAFC. Furthermore, the system efficiency of PAFCs are reported to be about 40% while that of MCFCs and SOFCs are expected to be in the high 40% range or 50-60%. An meaningful assessment of the cost of fuel cell systems is often difficult to obtain because many factors are used in the calculations, including the changing cost estimates associated with an improving technology as performance enhancements are realized. The PAFC systems probably offers the best opportunity to assess the capital costs for a fuel-cell technology that is ready for commercialization. The 4.5-MW demonstrator built by IFC, which was originally scheduled for test in New York City, had an estimated cost of about \$7800/kW (1978\$ US). In the early 1980s, an improved 4.5-MW PAFC plant was tested in Japan which had a capital cost of \$5560/kW (1980\$ US). The current generation of 11-MW PAFCs offered by IFC has an estimated purchase price of about \$3600/kW (1986\$ US) for the first three units, with the price of the next 20 units expected to be about \$1800/kW (1986\$ US). To date, no commitment has been made by US electric utilities to purchase any of the first three 11-MW PAFC units, however one unit that is designed for Japanese codes and standards will be operational at Goi, Japan in the early 1990s. A more recent study sponsored by the Electric Power Research Institute suggests that 25-MW PAFC systems with a heat rate of 8300 BTU/kWh (HHV) should have an installed cost of about \$1130/kW (1987\$ US). For a 100-MW PAFC plant operating at a lower heat rate (*i.e.*, higher efficiency), 7000 BTU/kWh (HHV), its capital cost is projected to be \$800/kW (1987\$ US).

Research and development of fuel-cell technology is progressing in the US. What is less certain is the progress in commercialization of fuel cells for terrestrial applications, particularly utility power generation. The PAFC technology is now available for purchase in 11-MW units from IFC, but as

yet no firm commitments from US utilities are evident. The primary impediments to commercialization of fuel cell technology appears to be: i) the unwillingness of manufacturers' to risk capital, ii) the reluctance of utilities to assume risks with new technology, and iii) the uncertainty of government and other institutions' willingness to support (subsidize) entry of new technology in the civilian market. If these issues are successfully resolved, then fuel cells have a good opportunity of becoming a viable technology for the future in the US.

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Table 1. Typical Fuel Cell Components

Type	Electrolyte	Temp. (°C)	Anode	Cathode	Bipolar Separator
AFC	75 wt% KOH	200	Ni	NiO	nickel
AFC	35-45 wt% KOH	80-90	Pt-20Pd	Pt-90Au	Au-plated Mg
AFC	35-45 wt% KOH	≤100	Pt/C	Pt/C	carbon-plastic composite
PEFC	ion-exchange membrane	≤100	Pt black pr Pt/C	Pt black or Pt/C	titanium or graphite
PAFC	100% H ₃ PO ₄	200	Pt/C	Pt/C	graphite
MCFC	62 mol% Li ₂ CO ₃ - 38 mol% K ₂ CO ₃	650	Ni-based material	Li-doped NiO	stainless steel alloys anode side Ni-plated
SOFC	Y ₂ O ₃ -ZrO ₂	1000	Ni-ZrO ₂ cermet	Sr-doped LaMnO ₃	Mg-doped LaCrO ₃ (cell interconnect)

Table 2. Typical Electrochemical Reactions in Fuel Cells

Fuel Cell	Anode Reaction	Cathode Reaction
Proton Exchange	$H_2 \rightarrow 2H^+ + 2e^-$	$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$
Alkaline	$H_2 + 2OH^- \rightarrow 2H_2O + 2e^-$	$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$
Phosphoric Acid	$H_2 \rightarrow 2H^+ + 2e^-$	$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$
Molten Carbonate	$H_2 + CO_3^{2-} \rightarrow H_2O + CO_2 + 2e^-$	$O_2 + 2CO_2 + 4e^- \rightarrow 2CO_3^{2-}$
Solid Oxide	$H_2 + O^{2-} \rightarrow H_2O + 2e^-$	$O_2 + 4e^- \rightarrow 2O^{2-}$

Table 3. Major Industrial Participants in United States Fuel Cell Technology

Industrial Participant	Location	Application/Activity
<i>Alkaline Fuel Cells</i>		
• IFC ^a	Connecticut	Space, military
<i>Polymer Electrolyte Fuel Cells</i>		
• Dow Chemical Co.	Texas	Polymer electrolyte R&D
• DuPont Inc.	Delaware	Polymer electrolyte R&D
• General Motors Corp.	Michigan	Transportation
• Treadwell Corp.	Connecticut	Military
• IFC	Connecticut	Transportation
• Ergenics Power Systems, Inc.	New Jersey	Portable power
<i>Phosphoric Acid Fuel Cells</i>		
• IFC	Connecticut	Utility power
• Westinghouse Electric Corp.	Pennsylvania	Utility power
• Energy Research Corp. (ERC)	Connecticut	R&D
<i>Molten Carbonate Fuel Cells</i>		
• Energy Research Corp. (ERC)	Connecticut	Internal-reforming fuel cell for utility power
• M-C Power	Illinois	Utility power
• IFC	Connecticut	Utility power
<i>Solid Oxide Fuel Cells</i>		
• Combustion Engineering	Connecticut	Monolith structure for utility power, military
• Garrett/AiResearch	California	Monolith structure for utility power, military
• IFC	Connecticut	Planar bipolar technology
• Ceramtec	Utah	Planar bipolar technology
• Westinghouse Electric Corp.	Pennsylvania	Tubular fuel cell for utility power
• Ztek Corp.	Massachusetts	Planar bipolar technology

^aInternational Fuel Cells Corp., subsidiary of United Technologies Corp.

Table 4. Major Fuel-Cell Stack Demonstrations by United States Developers

Manufacturer	Size/Type	Test Site/Application
• International Fuel Cells Corp.	12 kW/AFC	US Space Program
• International Fuel Cells Corp.	40 kW/PAFC	36 different sites
• International Fuel Cells Corp.	200 kW/PAFC	Japan
• International Fuel Cells Corp.	4.5 MW/PAFC	Japan
• Energy Research Corp.	5 kW/MCFC	in-house
• M-C Power	1 kW/MCFC	in-house
• International Fuel Cells Corp.	20 kW/MCFC	in-house
• Westinghouse Electric	3 kW/SOFC	Japan

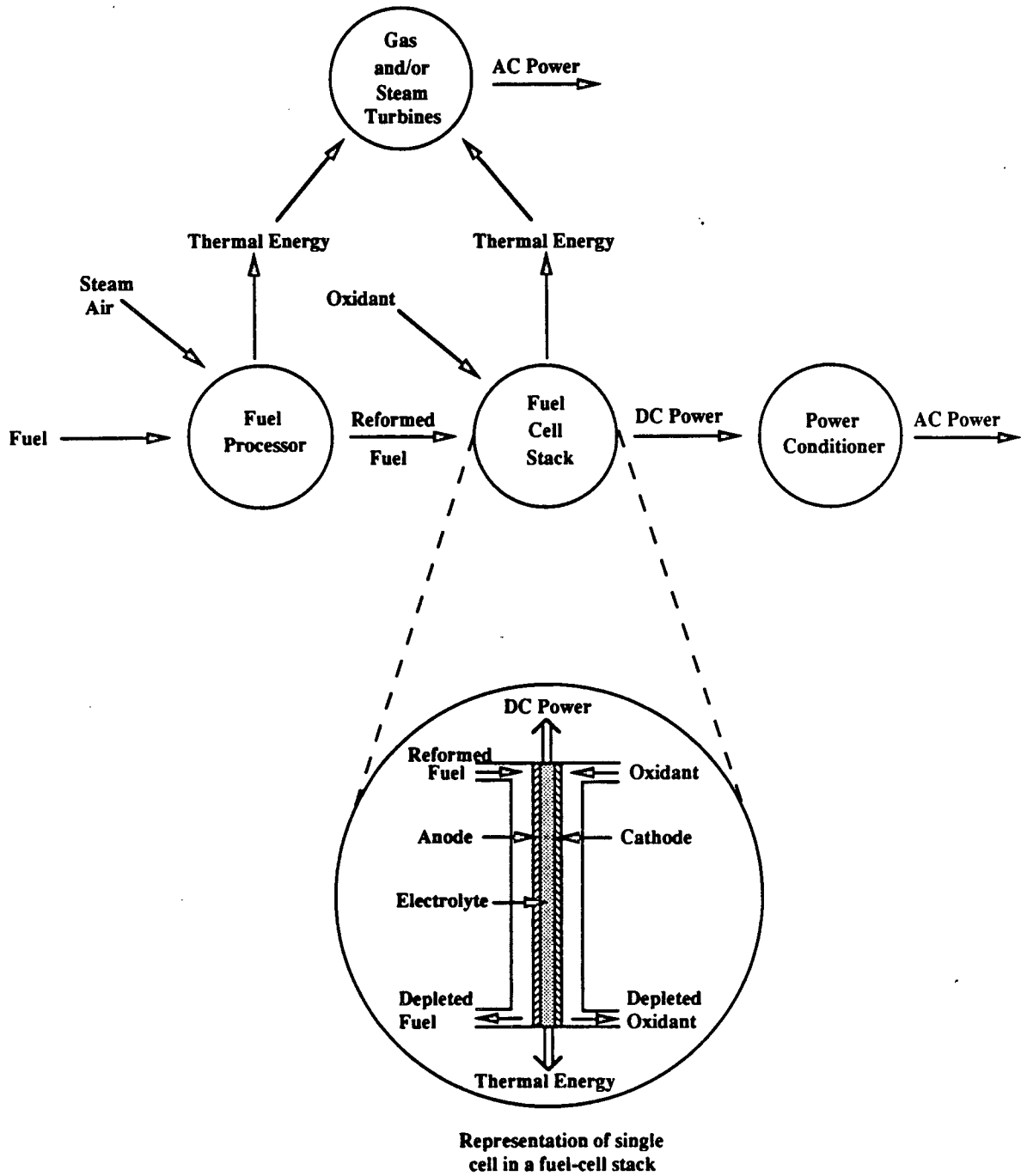


Figure 1. Schematic representation of fuel cell and integrated fuel cell plant consisting of fuel processor, fuel cell stack and power conditioner. Excess thermal energy is utilized to generate additional electric power by gas/steam turbines.

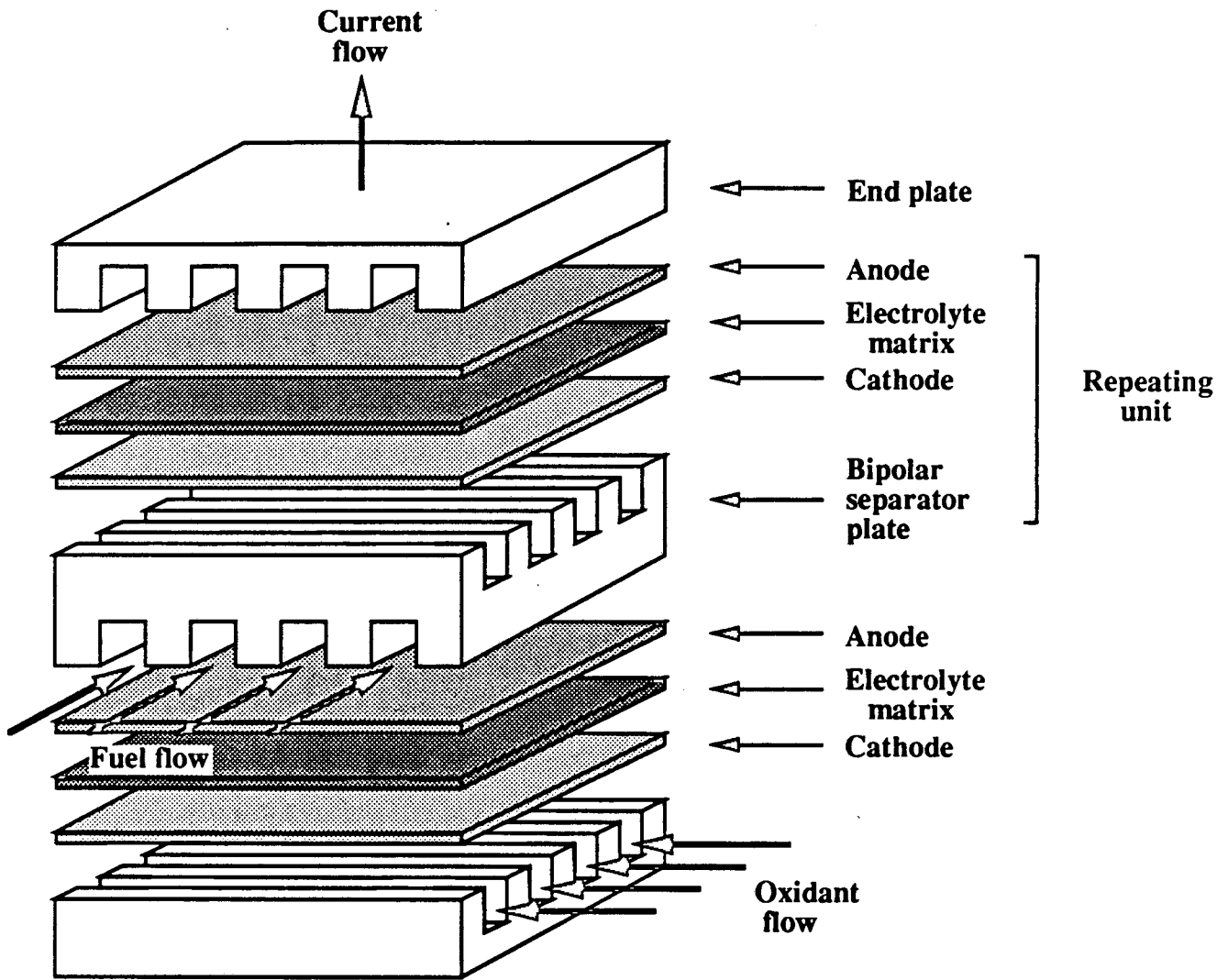


Figure 2. Schematic representation of cell components in a conventional bipolar stack arrangement with reactants in cross-flow pattern.

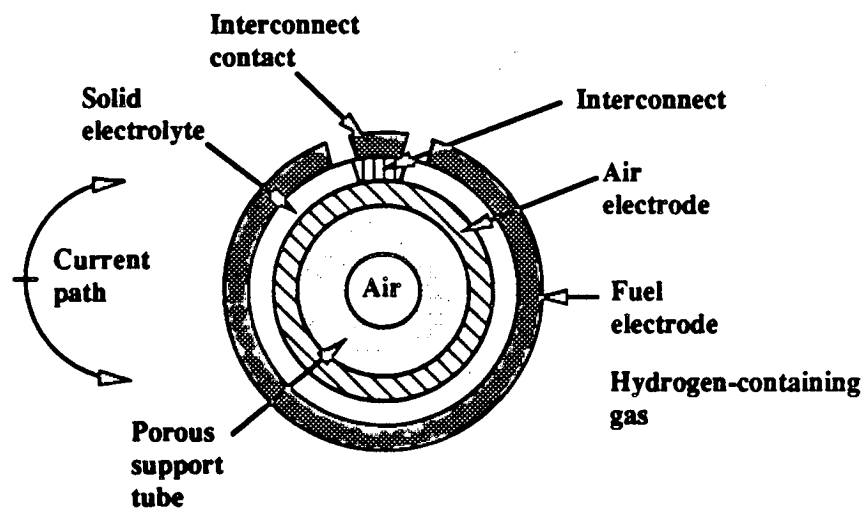


Figure 3. Cross-section view of tubular SOFC by Westinghouse Electric Corporation.

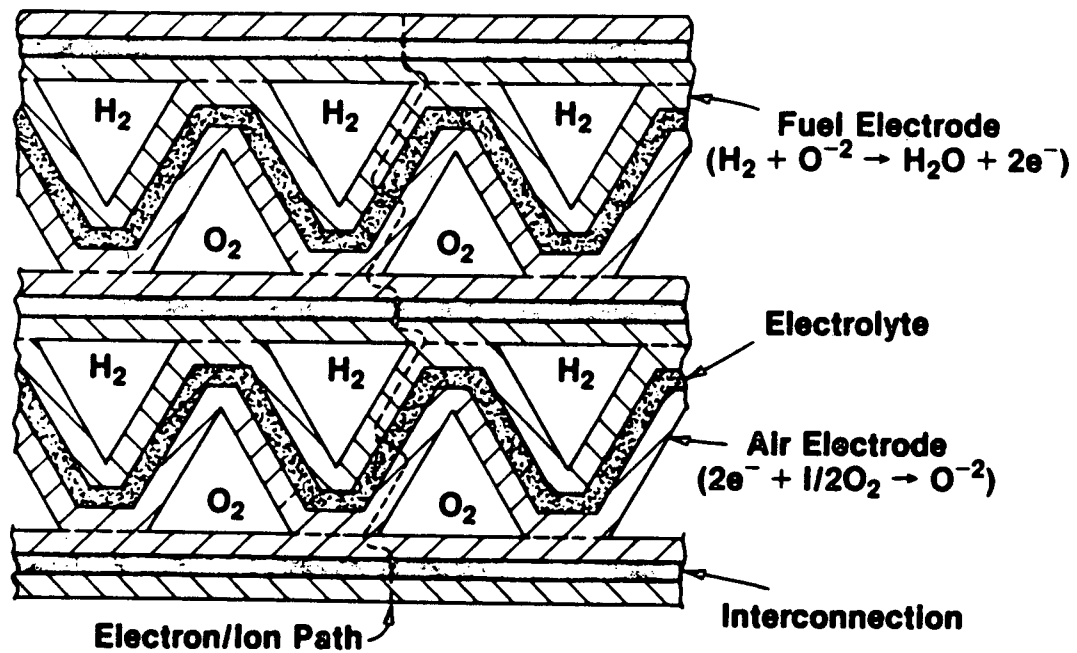


Figure 4. Schematic representation of monolith SOFC.

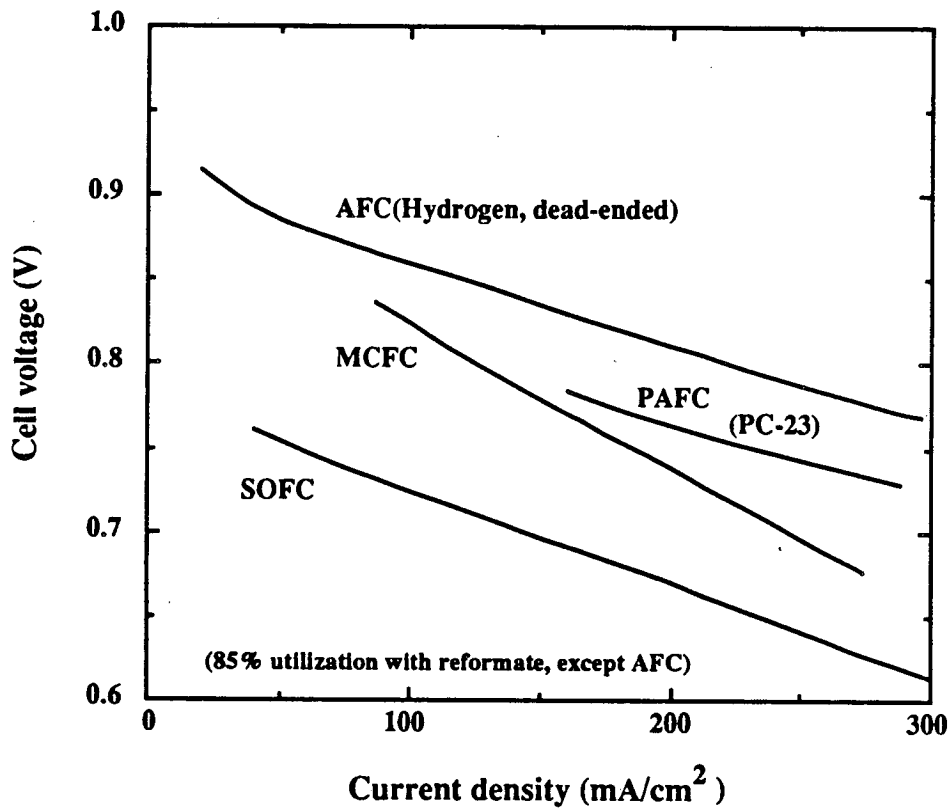


Figure 5. Examples of performance levels achievable with current technology in different types of fuel cells.

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