ELECTROCHEMICAL ELECTRODES

A fuel cell consists of a series of basic building blocks that incorporate an electrolyte layer in contact with a porous anode and porous cathode on either side. There are five main types of fuel cells: alkaline (AFC), proton exchange membrane (PEFC), phosphoric acid (PAFC), molten carbonate (MCFC), and solid oxide (SOFC). Each fuel cell uses different materials for all the components and, as a result, has different operating temperatures. In addition, some variations exist in the materials within each fuel cell type. However, the basic materials' properties are consistent within each fuel cell type.

A schematic of a typical fuel cell, along with the gas and ion conduction flow through the cell for PAFCs, PEFCs, MCFCs and SOFCs, is shown in Fig. 1. Ion specie and transport direction are different for each fuel cell type, which influences the site where water production will occur. In addition, each fuel cell type has different sensitivity to impurities and alternate fuels. The fuel and oxidant gases flow past the surface of the anode and cathode and generate electrical energy by the electrochemical oxidation of fuel, usually hydrogen, and the electrochemical reduction of oxygen. The electrolyte provides the environment for this reaction to occur. Flow of the fuel and oxidant gases is generally counter or cross flow rather than concurrent flow.

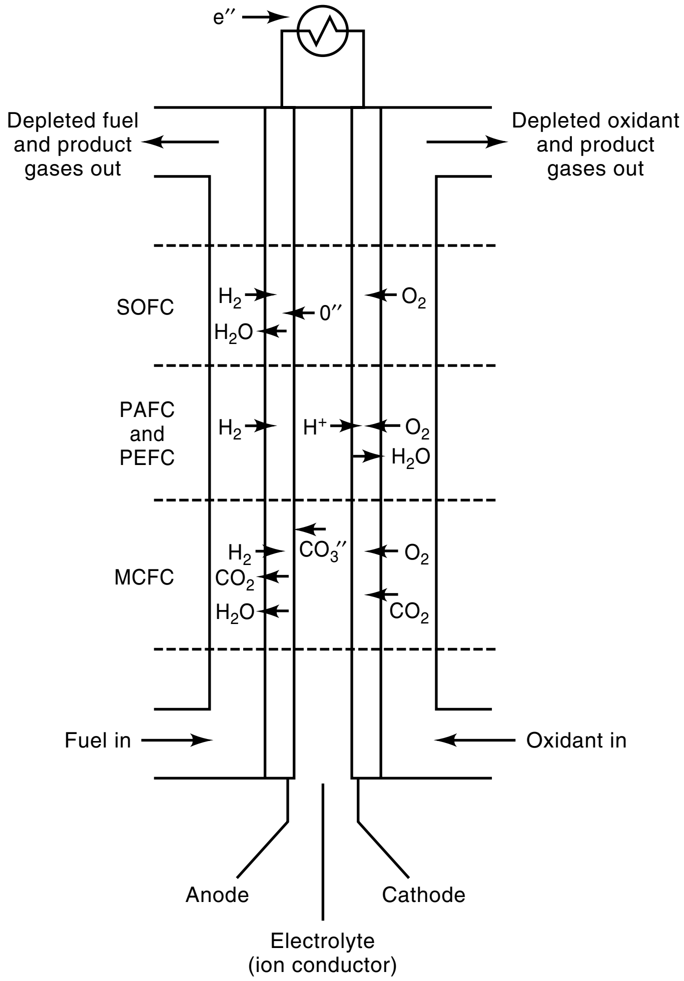

Figure 1. Schematic of typical gas and ion conduction flow through fuel cells.

The fuel can be any substance capable of being utilized through chemical oxidation (at the fuel cell operating temperature) that can be supplied continuously. PEFC's generally can only use hydrogen, but the higher temperature fuel types can utilize virtually any hydrogen rich stream (liquid or gaseous) and sometimes can utilize other materials compatible with the electrolyte such as carbon dioxide (CO_2) or ammonia (NH_3). Similarly, the oxidant can be any fluid or gas that can provide an oxygen-rich stream at the appropriate rate. However, the oxidant is generally air.

FUEL CELL COMPONENTS

The functions of electrodes in fuel cells are to: (1) provide a *surface* site where gas/liquid ionization of deionization reactions can take place, (2) *conduct* ions away from or into the three phase interface once they are formed (so an electrode must be made of materials that have good electrical conductance), and (3) provide a physical *barrier* that separates the bulk gas phase and the electrolyte (1). In order to provide the surface site and to increase the rates of reactions, the electrode material should be catalytic as well as conductive, porous rather than solid.

Ionization reaction rates increase with temperature. Therefore, the catalytic function of electrodes is more important in lower temperature fuel cells and less so in high temperature fuel cells. In addition, the porous electrodes must be permeable to both electrolyte and gases, but not such that the media can be easily "flooded" by the electrolyte or "dried" by the gases in a one-sided manner.

In addition to transporting dissolved reactants to the electrode, the electrolyte also conducts ionic charge between the electrodes and thereby completes the cell electric circuit, as illustrated in Fig. 1. It also provides a physical barrier to separate the fuel and oxidant gas steams. In low temperature fuel cells (PEFC, AFC, PAFC), protons or hydroxyl ions are the major charge carriers in the electrolyte, whereas in the high temperature fuel cells, MCFC and SOFC, carbonate ions and oxygen ions are the charge carriers, respectively.

Individual fuel cell packages or sandwiches (anode, electrolyte, and cathode) generally do not provide sufficient power for most applications. Therefore, individual fuel cells must be combined to produce appreciable amounts of electricity and are joined by interconnects. Because of the configuration of a flat plate cell, Fig. 2, the interconnect becomes a separator plate which has two functions: (1) to provide an electrical series connection between adjacent cells, and specifically for flat plate cells, and (2) to provide a gas barrier that separates the fuel and oxidant of adjacent cells. The interconnect of the tubular solid oxide fuel cell is a special case and has a slightly altered function. All interconnects must be an electrically conducting and impermeable to gases.

Depending on the type of fuel cell, other parts of the cell could include (1) the structure for distributing the reactant gases across the electrode surface and which serve as mechanical support, shown as ribs in Fig. 2, (2) electrolyte reservoirs for liquid electrolyte cells to replenish electrolyte lost over life, and (3) current collectors (not shown) which provide

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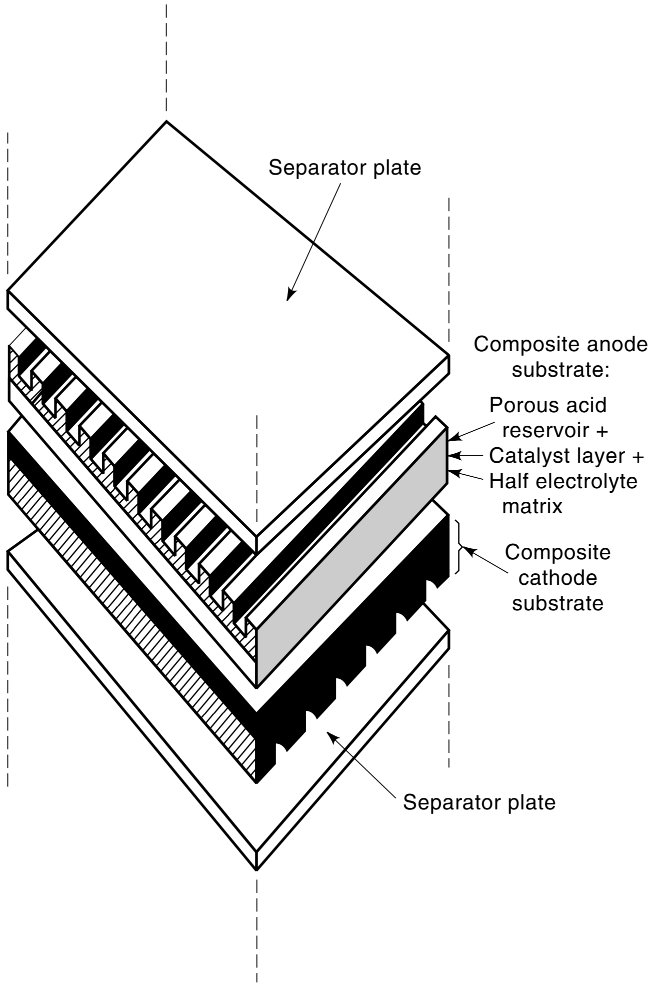

Figure 2. Typical planar fuel cell configuration.

a path for the current between the electrodes and the separator of flat plate cells.

The physicochemical and thermomechanical properties of materials used in the cell components (i.e., electrodes, electrolyte, interconnect, current collector, etc.) determine the practical operating temperature and useful life of a fuel cell. Aqueous electrolytes are limited to temperatures of about 200°C or lower because of their high water vapor pressure and/or rapid degradation at higher temperatures. The operating temperature also plays an important role in dictating the type of fuel that can be utilized in a fuel cell. The low temperature fuel cells with aqueous electrolytes are, in most practical applications, restricted to hydrogen (which includes reformed methane) as a fuel. In high temperature fuel cells, carbon monoxide (CO) and even methane (CH_4) can be used directly because of the inherently rapid electrode kinetics and the lesser need for high electrocatalytic activity at high temperature (2).

THE ELECTRODE ROLE

The chemical and physical interactions that occur at the electrode interface are critical to the performance of the fuel cell. A three-phase interface is established in the region of the porous electrodes, the electrolyte, and the gaseous reactants. The nature of this interface 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 an electrolyte film that wets portions of the porous electrode and react electrochemically on the electrode surface. The balance of fuel cell and open porosity is critical to the performance of the fuel cell. 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.

Much of the recent effort in the development of fuel cell technology has been devoted to reducing the thickness of cell components while refining and improving the electrode structure and the electrolyte phase, with the aim of obtaining a higher and more stable electrochemical performance, while simultaneously reducing the cost of the materials and system.

Electrode Reactions

In a fuel cell, the anodic and cathodic reactants are fed into their respective chambers, and an electrolyte layer provides a region for the gases to move through the system. The half cell reaction at the anode yields electrons, shown in Table 1. These electrons are transported through the external circuit and reach the cathode where they are transferred to the cathodic reactants. The cycle is completed by the transport of ions from one electrode to the other through the electrolyte.

The typical electrochemical reactions that occur with different fuels and oxidants for the various fuel cell types are also summarized in Table 1. CO and CH_4 are shown in the table as undergoing anodic oxidation, but in most cases, direct oxidation may not occur. Instead, these reactants are potential fuels because they undergo chemical reaction with water (H_2O) to produce hydrogen (H_2), which is the oxidizable fuel. In MCFCs, CO, and CH_4, combined with H_2O in the proper environment, are sources of H_2 from water gas shift and stream reforming reactions, respectively. The direct oxidation of CO and CH_4 in high temperature SOFCs is feasible because of the high temperature.

Low temperature fuel cells (AFC, PEFC, PAFC) require separate reformers containing noble metal electrocatalysts to achieve practical reaction rates at the anode and cathode, and hydrogen is the only acceptable fuel. Impurities in the fuel rapidly decrease the performance of the system. Carbon monoxide "poisons" the noble metal anode electrocatalyst such as platinum (Pt) in low temperature fuel cells. However, with high temperature fuel cells (MCFC, SOFC), the requirements for electrocatalysis are relaxed, and the number of potential fuels is increased. Carbon monoxide serves as a potential source of H_2 in high temperature fuel cells where nonnoble metal electrocatalysts such as nickel (Ni) are used.

The overall electrochemical reactions corresponding to the individual electrode reactions listed in Table 1 along with the appropriate form of the Nernst equation, are shown in Table 2. The Nernst equation provides a relationship between the standard potential (E^0) for the cell reaction and the equilibrium potential (E) at various temperatures and partial pressures (activities) of reactants and products. According to the Nernst equation, the equilibrium cell potential at a given temperature can be increased by operating at higher reactant

Table 1. Typical Electrochemical Reactions in Fuel Cells

Fuel Cell	Anode Reaction	Cathode Reaction
Proton exchange	$H_2 \rightarrow 2H^+ + 2e^-$	$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$
Alkaline	$H_2 + 2(OH) \rightarrow 2H_2O + 2e^-$	$\frac{1}{2}O_2 + H_2O + 2e^- \rightarrow 2(OH)$
Phosphoric acid	$H_2 \rightarrow 2H^+ + 2e^-$	$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$
Molten carbonate	$H_2 + CO_3^= \rightarrow H_2O + CO_2 + 2e^-$ $CO + CO_3^= \rightarrow 2CO_2 + 2e^-$	$\frac{1}{2}O_2 + CO_2 + 2e^- \rightarrow CO_3^=$
Solid oxide	$H_2 + O^= \rightarrow H_2O + 2e^-$ $CO + O^= \rightarrow CO_2 + 2e^-$ $CH_4 + 4O^= \rightarrow 2H_2O + CO_2 + 8e^-$	$\frac{1}{2}O_2 + 2e^- \rightarrow O^=$

pressures. In fact, improvements in fuel cell performance have been observed at higher pressures.

The fuel (generally hydrogen) enters the pores of the anode and reaches the reaction zone where gas, the electrolyte, and the solid conducting structure meet. This is the "three-phase interface." Hydrogen diffuses to the electrochemically active site where it is adsorbed and dissolved by the electrolyte. Dissociation and ionization then occur.

The electrochemical reactions of H_2 and O_2 in fuel cells produce H_2O. When a carbon containing fuel is involved in the anode reaction, CO_2 is also produced. In the case of the MCFC, CO_2 is required in the cathode reaction to maintain an invariant carbonate concentration in the electrolyte. Since CO_2 is produced at the anode and consumed at the cathode in MCFCs, and the concentrations in the anode and cathode feed streams are not necessarily equal, the Nernst equation in Table 1 includes the CO_2 partial pressure for both electrode reactions.

In some cases, the validity of the mechanism depends on the surface structure of the electrodes and the current operating conditions. Cell potential/current density and charge curves allow determination of the mechanism. The data for these special potentiostatic and potentiodynamic analyses can be obtained on rotating and stationary ring-disc electrodes, as well as impulse methods.

Porous Gas Electrodes

Since reactants for the fuel cells are usually gaseous, the most significant breakthrough in fuel cell technology has been the development of porous gas diffusion electrodes. The higher temperature fuel cells can provide power densities greater than those produced in combustion engines.

The principle function of the gas diffusion electrode is to provide a large reaction zone area with a minimum of mass transport resistance (for the reactants and removal of byproducts). Since electrode potential changes with the current density (current per unit area), a substantial increase in electrode area has an important effect on cell performance. Large three-phase zones can be achieved by the use of metal powders with a very high specific area (>100 m^2/g) or carbon (1000 m^2/g).

Surface Area Effect. Porous electrodes can have surface areas which are orders of magnitude larger than their geometric area. Fillers and binders are sometimes mixed with the electrode components during manufacture and removed at the end of the manufacturing process. This leaves voids that enhance the porosity of the structure and permit the electrode to have a hydrophobic or hydrophilic surface. Hydrophobic electrodes are made of fine carbon powder bonded with a plastic material. The porous electrodes that are used in low temperature fuel cells consist of a composite structure that contains Pt electrocatalyst on a high surface area carbon black and a polytetrafluoroethylene (PTFE) binder. Carbon powders are light, have a large surface area, are suitable for the deposition of active catalysts, such as platinum, and are readily manufactured on a commercial scale. Such electrodes for acid and alkaline fuel cells are described by Kordesch et al. (3). In these porous electrodes, PTFE is hydrophobic (acts as a "wet-proofing" agent) and serves as the gas permeable phase. In addition, carbon black is an electron conductor that provides a high surface area to support the electrocatalyst. Platinum serves as the electrocatalyst which promotes the rate of electrochemical reactions (ionization/deionization) for a given surface area. The carbon black also has a certain degree of hydrophobicity, depending on the surface properties of the material.

Two layers can be identified in these PTFE bonded carbon electrodes: a highly hydrophobic porous gas diffusion layer and an electrolyte-wettable thin layer. The composite structure of PTFE and carbon establishes an extensive three phase interface in the porous electrode, which is the benchmark of

Table 2. Nernst Equations for Fuel Cell Reactions[a]

Cell Reaction	Nernst Equation
$H_2 + \frac{1}{2}O_2 + \rightarrow H_2O$	$E = E^0 + (RT/2F)\ln[P_{H_2}/P_{H_2O}] + (RT/2F)\ln[P_{O_2}^{1/2}]$
$H_2 + \frac{1}{2}O_2 + CO_2(c) \rightarrow$ $H_2O + CO_2(a)$	$E = E^0 + (RT/2F)\ln[P_{H_2}/P_{H_2O}(P_{CO_2})_a]$ $+ (RT/2F)\ln[P_{O_2}^{1/2}](P_{CO_2})_c]$
$CO + \frac{1}{2}O_2 \rightarrow CO_2$	$E = E^0 + (RT/2F)\ln[P_{CO}/P_{CO_2}] + (RT/2F)\ln[P_{O_2}^{1/2}]$
$CH_4 + 2O_2 \rightarrow 2H_2O + CO_2$	$E = E^0 + (RT/8F)\ln[P_{CH_4}/P_{H_2O}^2 P_{CO_2}]$ $+ (RT/8F)\ln[P_{O_2}^2]$

[a] (a) = anode; (c) = cathode; E = equilibrium potential; E^0 = standard potential; P = partial pressure of gas; R = universal gas constant; T = temperature.

PTFE bonded electrodes. The electrochemical reaction takes place at the interface, where the catalytically active material must be present.

It should be noted that the conductivity of carbon is not sufficient for high currents (>200 mA/cm^2). To improve their conductivity, a wire screen as a current collector or a bipolar plate material is required.

Sintered metal powders are used for hydrophilic electrodes. The gas diffusion layer of these electrodes has larger pores than the reaction layer. Although these electrodes are heavy, they have a high conductivity, which is a great advantage in monopolar plate electrodes, where the current is collected from tabs at the edges of the electrodes.

Current Density. The current densities that are obtained from smooth electrodes are usually in the range of single units mA/cm^2 or less because of rate limiting processes such as the available area of the reaction sites. Porous electrodes are commonly used in fuel cells to achieve much higher current densities. These high current densities are possible because the electrode has a high surface area which significantly increases the amount of reaction sites, and the optimized electrode structure has favorable mass transport properties. In an idealized porous gas fuel cell electrode, high current densities at reasonable polarization are obtained when the liquid (electrolyte) layer on the electrode surface is sufficiently thin so that it does not significantly impede the transport of reactants to the electroactive sites, and a stable three phase (gas/electrolyte/electrode surface) interface is established.

In MCFCs, which operate at relatively high temperatures, there are no known materials that can serve to wet proof a porous structure against permeation by molten carbonates. Consequently, the technology used to obtain a stable, three phase interface in MCFC porous electrodes is different from that used in PAFCs. In the MCFC, the stable interface is achieved in the electrodes by carefully tailoring the pore structures of the electrodes and the electrolyte matrix ($LiAlO_2$) so that the capillary forces establish a dynamic equilibrium in the different porous structures.

SOFCs, on the other hand, do not have a liquid electrolyte present that is susceptible to movement in the porous electrode structure, and electrode flooding is not a problem. Consequently, the three phase interface that is necessary for efficient electrochemical reaction involves two solid phases (solid electrolyte/electrode) and a gas phase. A critical requirement of porous electrodes for SOFC is that they are sufficiently thin and porous to provide an extensive electrode/electrolyte interfacial region for electrochemical reaction.

Transport Processes

The transport processes involving the mass transfer of reactants/products play a prominent role in the performance of porous electrodes in fuel cells. Transport processes involving heat transfer, and thermal management are important in fuel cell systems. The slow transport of reactant and product species through the porous electrode gives rise to the polarization. A sequence of steps, involving both transport and rate processes, occurs when a reactant species undergoes electrochemical reaction in a porous electrode. One sequence suggested by Liebhafsky and Cairns (4) indicates there are 13 steps involved in the overall electrochemical reaction in a porous fuel cell electrode. With the exception of the dissolution of the reactant in the electrolyte and the transport of the reactant through the electrolyte to the electrode surface, each of the other steps can be associated with a polarization that could contribute to an increase in the inefficiency of the electrode reaction. These include physical, chemical, and electrochemical reactions in the fuel cell. Not all of the steps contribute significantly to the total polarization, and those steps can be disregarded. Thus, efforts to improve the performance of porous fuel cell electrodes can be directed at reducing the polarization associated with adsorption of the electroactive species onto the electrode, the surface migration of the absorbed species, the electrochemical reaction of the electrically charged species, and the transport of gaseous and liquid products away from the surface and interface.

Transport processes involving diffusion, convection, and migration can take place in the gas phase in the pores of the electrodes or in the liquid phase in the pores of the electrode. The relative contribution of these transport processes to the overall electrode polarization is affected by the porous electrode structure, electrolyte composition and temperature, and reactant gas composition and pressure.

The maximum rate of transport of reactant species to the electrode surface provides an upper limit to the rate of electrochemical reaction. Under these conditions, a limiting current is reached. In a fuel cell with porous electrodes containing a liquid (electrolyte) layer, the diffusion of the reactant species through the electrolyte is usually the rate limiting transport process.

Irreversible Thermodynamics. Useful amounts of work (electrical energy) are obtained from a fuel cell only when a reasonably large current is drawn, but the cell potential will be decreased from its equilibrium potential because of irreversible losses. There are several sources that contribute to irreversible losses in a practical fuel cell. The losses, which are often called polarization, overpotential, or overvoltage, originate primarily from three sources: (1) ohmic polarization (η_{ohm}), (2) concentration polarization (η_{conc}), and (3) activation polarization (η_{act}). These losses result in a cell voltage (V) for a fuel cell that is less than its reversible potential.

Ohmic Polarization. The ohmic losses occur because of resistance to the flow of ions in the electrolyte and resistance to flow of electrons through the electrode materials. The dominant ohmic losses through the electrolyte phase are reduced by decreasing the electrode separation and enhancing the ionic conductivity of the electrolyte. Because both the electrolyte and fuel cell electrodes obey Ohm's law, the ohmic losses can be expressed by the equation,

$$\eta_{\mathrm{ohm}} = iR$$

where i is the current flowing through the cell, and R is the total cell resistance, which includes electronic, ionic and contact resistances.

Concentration Polarization. As a reactant is rapidly consumed at the electrode by electrochemical reaction, concentration gradients will be established. Several processes may contribute to concentration polarization: (1) slow diffusion in the gas phase in the electrode pores, (2) solution/dissolution of reactants/products into/out of the electrolyte, and/or (3) diffusion of reactants/products through the electrolyte to/from the electrochemical reaction site. At practical current densities, slow transport of reactants/products to/from the electrochemical reaction site is a major contributor to concentration polarization.

Activation Polarization. Activation polarization is present when the rate of an electrochemical reaction at an electrode surface is associated with sluggish electrode kinetics. In other words, activation polarization is directly related to the rates of electrochemical reactions. There is a close similarity between electrochemical and chemical reactions in that both involve an activation barrier that must be overcome by the reacting species. Processes involving absorption of reactant species, transfer of electrons across the double layer, desorption of product species, and the nature of the electrode surface can all contribute to activation polarization.

Electrode Polarization. Activation and concentration polarizations can exist at both the positive (cathode) and negative (anode) electrodes in fuel cells. The net result of current flow in a fuel cell is to increase the anode potential and to decrease the cathode potential, thereby reducing the cell voltage. The cell voltage includes the contribution of the polarization and the anode and cathode potentials. Current flow in a fuel cell results in a decrease in the cell voltage because of losses by electrode and ohmic polarizations. The goal of fuel cell developers is to minimize the polarization so that the cell voltage (V_{cell}) approaches the difference between the anode and cathode potential (ΔE_e). This goal is approached by modifications to the fuel cell operating conditions (i.e., higher gas pressure, higher temperature, change in gas composition to lower the gas impurity concentration, etc.), improvement in electrode structures, better electrocatalysts, more conductive electrolyte, etc. However, for any fuel cell, trade-offs exist between achieving higher performance by operating at higher temperature or pressure and the problems associated with the stability/durability of cell components encountered at the more severe conditions.

Although ohmic, concentration, and activation polarization are the classic fuel cell losses, others exist in the electrodes. Two worthy of consideration are contact resistant losses and losses due to exchange currents.

Contact Resistance. Contact resistances at electrodes and separators account for losses due to spatial variation in material properties, fluxes, reaction rate densities, etc. Contact or terminal resistance is actually a form of ohmic resistance.

Exchange Current. This is the exchange of electrons through the electrolyte rather than through the external load. This is the only form of losses that decreases with an increase in current output. Exchange current is directly related to the chemical potential difference, so even at zero external load current, there are electrons delivered to the cathode. Once oxygen ions are formed, they migrate through the electrolyte to the anode where they deionize to release an electron. The electron released migrates back to the cathode to continue the process or "exchange." With the ionization/deionization reactions proceeding at a low (but non-zero equilibrium) rate, the cell voltage is diminished below that of reversible potential, even though there is no external current flowing. This form of loss, however, would become less significant once the external current is increased beyond a certain value. Moreover, the magnitude of the exchange current actually decreases with an increase in external current since the cell voltage, which drives the occurrence of exchange current, would decrease further.

POLYMER ELECTROLYTE FUEL CELL ELECTRODES

The electrolyte in this fuel cell is an ion exchange membrane (fluorinated sulfonic acid polymer or other similar polymers) which is an excellent proton conductor. The only liquid in this fuel cell is water; thus, corrosion problems are minimal. Water management in the membrane is critical for efficient performance; the fuel cell must operate under conditions where the byproduct water does not evaporate faster than it is produced because the membrane must be hydrated. Because of the limitation on the operating temperature imposed by the polymer and problems with water balance, usually less than 120°C, a H_2-rich gas with little or no CO is used, and higher catalysts loadings (Pt in most cases) than those used in PAFCs are required in both the anode and cathode.

The electrodes for this system are typical gas-diffusion electrodes. The backing is a porous carbon cloth with a hydrophobic coating. In order for efficient electrochemical reactions to take place, the electrodes must be catalyzed. Platinum is generally considered the best catalyst for both the hydrogen oxidation and the oxygen-reduction reactions. To reduce the amount of catalyst required and increase the surface area, the use of supported platinum catalysts, similar to those used in the phosphoric acid cell, have been adopted. These consist of 2 to 5 nm diameter platinum particles on the surface of fine carbon particles. Unfortunately, Ticianelli et al. (5) determined that only about 10 to 20% on the platinum is electrochemically active in the fuel cell reaction. Research efforts are focused on increasing the catalyst activity.

To function, the catalyst must contact the gas as well as both the electrical and protonic conductors. This is done by impregnating the supported-catalyst electrode with protonic conducting material. The most common method is to cover the surface with a solution of solubilized membrane material of Nafion or its equivalent. Newly developed materials have resulted in catalyst loadings down to 0.25 mg $\mathrm{Pt/cm^2}$ (with 0.716 V), which greatly improves the commercial viability of this concept.

ALKALINE FUEL CELL ELECTRODES

The first successful, commercially utilized fuel cell was the H_2-O_2 alkaline cell, which was used in the space program. However, this system has found little other commercial applications.

The electrolyte in this fuel cell is concentrated potassium hydroxide (85 wt% KOH) in fuel cells operated at 250°C or less concentrated (35 to 50 wt%) KOH for lower temperature ($<$120°C) operation. The electrolyte is retained in a matrix (usually asbestos). The thermodynamics of this system and the use of pure hydrogen as the fuel, permit the use of a wide range of electrocatalysts (e.g., Ni, Ag, metal oxides, spinels, and noble metals).

The alkaline fuel cells used for the space shuttle orbiter are a significantly advanced verison of the early Bacon cells and the Apollo service module. They were developed by a Pratt and Whitney Corporation division which is now known as International Fuel Cells. The early orbiter module operated at a pressure of 4.0×10^5 to 4.4×10^5 N/m^2 and a temperature of about 90°C. The high performance at low temperature was achieved using Teflon-bonded electrodes with pure, high-loading noble metal catalysts. The anode consisted of pure noble metal black (80% platinum, 20% palladium) on a silver-plated nickel screen. The cathode had 90% gold with 10% platinum on a gold-plated nickel screen. Later versions, which are proprietary, of this system had improved materials, different operating parameters, less noble metal loading, and electrolyte reservoirs. However, this system is still far from being cost-effective for everyday applications (2).

More commonly, the systems are built with catalyzed carbon on nickel screen current collectors or porous nickel substrates with a hydrophobic backing. Platinum is the key ingredient in the catalyst. Because these systems operate at such low temperatures, plastics can be used for the cell frames.

More recent advances involve the use of a porous gas diffusion layer with 60 wt% PTFE on carbon, a nickel wire mesh, and an active layer of 12.24 wt% cobalt tetraphenylporphyrin (CoTPP) and 12 wt% PTFE on carbon for the cathode. The catalyst for the anode consists of 8.3 wt% platinum, 8.3 wt% palladium and 17 wt% PTFE on charcoal (6). Additional research is being conducted to improve the catalyst and electrodes (7).

PHOSPHORIC ACID FUEL CELL ELECTRODES

The phosphoric acid fuel cell is commercially available today in both stationary and on-site cogeneration plants. International Fuel Cells Corporation/ONSI is the recognized technology leader in this field and manufactures the only commercially available units. (These units are also sold by Toshiba and Ansaldo.) The electrochemical reactions for this system occur on highly dispersed electrocatalyst particles supported with carbon black. Catalysts are used on both the anode (Pt) and cathode (Pt or an alloy containing Pt).

In the early 1960s, the conventional porous electrodes were PTFE-bonded Pt-black, and the platinum loadings were extremely high—about 9 mg Pt/cm^2. The hardware was gold-plated Tantalum. During the past two decades, the cost of the materials and the requirement for Pt has decreased. A major breakthrough was the development of carbon blacks and graphite for cell construction materials (8,9). At that time, it was shown that carbon black and graphite were sufficiently stable to replace the more expensive gold-plated tantalum cell hardware. Because the carbon black had high surface areas, a dramatic reduction in Pt loading could be obtained with little or no reduction in performance. Appleby (8) reports that "without carbon, a reasonably inexpensive acid fuel cell would be impossible, since no other material combines the necessary properties of electronic conductivity, good corrosion resistance, low density, surface properties (especially in high-area form) and above all, low cost." Table 3 shows the evolution of the cell component technology for these fuel cells.

Concentrated phosphoric acid is used for the electrolyte in this fuel cell, which operates between 150 and 220°C. At lower temperatures, phosphoric acid is a poor ionic conductor, and CO poisoning of the Pt electrocatalyst in the anode becomes more severe. The relative stability of concentrated phosphoric acid is high compared to other common acids; consequently, the PAFC is capable of operating at the high end of the acid temperature range (100 to 220°C). In addition, the use of concentrated acid ($\approx$100%) minimizes the water vapor pressure, so water management in the cell is not difficult. The matrix universally used to retain the acid is silicon carbide (10), and the electrocatalyst in both the anode and cathode is Pt.

The porous electrodes used in this system are described extensively in the patent literature (11). These electrodes contain a mixture of the electrocatalyst supported on carbon black and a polymeric binder such as PTFE (30 to 40 wt%). The polymer binds the carbon black particles together to form an integral porous structure. A carbon paper substrate is also used and serves a dual role as a structural support for the electrocatalyst layer as well as the current collector. A typical carbon paper used in PAFCs has an initial porosity of about 90%. This wet-proof carbon paper should contain macropores of 3 to 50 μm diameter, with a median pore diameter of about 12.5 μm. It should also have micropores with a median pore diameter of about 34 Å for gas permeability. The composite structure consisting of a carbon black/PTFE layer on carbon-paper substrate forms a stable, three-phase interface in the fuel cell, with H_3PO_4 electrolyte on the electrocatalyst side and the reactant gas environment on the other side of the carbon paper (12).

The individual cells are physically separated by a bipolar plate which also serves to connect the cells electrically in the fuel cell stack. This plate also contains the gas channels for introducing the reactant gases to the porous electrodes and removing the products and inerts. Heat-treated bipolar plates are made from carbonized graphite-resin plates. The heat treatment, which occurs at about 2700°C, improves the corrosion resistance. That is, the corrosion current is reduced by two orders of magnitude at 0.8 V in 97% H_3PO_4 at 190°C and 486 kPa. This is important for the goal of 40,000 h of life for the stack (13).

Electrode performance may degrade rapidly by flooding due to carbon corrosion and loss of PTFE. This is sometimes observed in cells with high initial performance. On the contrary, an electrode having low initial performance may have long life; it has high wetproofing ability to the electrolyte, which improves the formation of the three-phase boundary (14).

Despite the progress in the development of cell components over the past decade, there is still significant research underway on alternative and component improvements that have the potential of cost reductions and performance improvements. This includes improved catalysts, catalyst supports, and electrolyte management. Alternative support materials with improved physicochemical properties are desired to increase the flexibility and remove the current temperature and electrical constraints on PAFCs. A large number of materials offer potential (15).

MOLTEN CARBONATE FUEL CELL ELECTRODES

MCFC is an intermediate temperature fuel cell that operates between 600 and 700°C. The chronology of component development is shown in Table 4. Since the mid-1970s, the materials for the electrodes and electrolyte structure have remained essentially unchanged. The primary developments have been the modification of the electrolyte and improvements in the

Table 3. Evolution of Phosphoric Acid Fuel Cell Technology

Component	1965	1975	Current
Anode	PTFE-bonded Pt black	PTFE-bonded Pt/C	PTFE-bonded Pt/C
		Furnace black	Furnace black
Anode catalyst	9 mg/cm^2 Pt	0.25 mg Pt/cm^2	0.25 mg Pt/cm^2
Cathode	PTFE-bonded Pt black	PTFE-bonded Pt/C	PTFE-bonded Pt/C
		Furnace black	Furnace black
Cathode catalyst	9 mg/cm^2 Pt	0.5 mg Pt/cm^2	0.5 mg Pt/cm^2
Electrode support	Ta mesh screen	Carbon paper	Carbon paper
Electrolyte support	Glass fiber paper	PTFE-bonded SiC	PTFE-bonded SiC
Electrolyte	85% H_3PO_4	95% H_3PO_4	100% H_3PO_4

fabrication processes. Developments in the fuel cell components have been reviewed by many people including Maru (16,17), Petri (18), Selman (19), and others.

The electrolyte in this fuel cell is usually a combination of alkali (Li, Na, K) carbonates, which is retained in a ceramic matrix of $LiAlO_2$. The fuel cell operates where the alkali carbonates form a highly conductive molten salt, with carbonate ions providing ionic conduction. At the high operating temperatures in MCFCs, Ni (anodes) and nickel oxide (cathodes) are adequate to promote reaction, and noble metals are not required. However, the major problems with Ni-based anodes and NiO cathodes are structural stability and NiO dissolution. The dissolution of the NiO in molten carbonate electrolyte became evident when thin electrolyte layers were used. Although NiO has a low solubility (10 ppm) in the electrolyte, Ni ions diffuse in the electrolyte toward the anode, and metallic Ni can precipitate in the reducing environment by the anode. This phenomenon becomes worse at high CO_2 partial pressures (20,21) because dissolution may involve the following mechanism:

$$NiO + CO_2 \rightarrow Ni^{2+} + CO_2^{2-}$$

The dissolution of NiO has been correlated to the acid-base properties of the electrolyte. The basicity of the electrolyte is directly proportional to the CO_2 partial pressure ($\log P_{CO_2}$). In addition, the solubility of NiO in the electrolytes is complicated by its dependence on several additional parameters: carbonate composition, H_2O partial pressure, and temperature.

The bipolar plates used in MCFC stacks are usually fabricated from thin sheets of an alloy (such as Incoloy 825, 310S, or 316L stainless steel) that is coated on the inlet side with Ni. The Ni layer is stable in the reducing gas environment of the anode compartment and provides a conductive surface coating with low contact resistance. To minimize corrosion at the wet-seal area, a thin Al coating is usually applied. This layer reacts with the Al and electrolyte and becomes $LiAlO_2$.

Table 4. Evolution of Molten Carbonate Fuel Cell Technology

Component	1965	1975	Current
Anode	Pt, Pd, or Ni	Ni-10 wt% Cr	Ni-10 wt% Cr 3–6 μm pore 50–60% porosity 0.5–1.5 mm thick 0.1–1 m^2/g
Cathode	Ag_2O or lithiated NiO	Lithiated NiO	Lithiated NiO 7–15 μm pore 70–80% porosity 0.5–0.75 mm thick 0.5 m^2/g
Electrolyte support	MgO	Mixture of α-, β-, and γ-$LiAlO_2$ 10–20 m^2/g	γ-$LiAlO_2$ 0.1–12 m^2/g
Electrolyte	52% Li, 48% Na 43.5% Li, 31.5% Na, 25% K	62% Li, 38% K 60–65 wt% Li	62% Li, 32% K 50% Li, 50% Na 50% Li, 50% K 50 wt% Li
Fabrication process	Paste	Hot press “tile” 1.8 mm thick	Tape-cast Dry press Printing process 0.5 mm thick

However, it is important that $LiAlO_2$ not be present in those areas where electronic conduction is required since this is an insulating material. Thus, it is not suitable for providing corrosion resistance to the cell current collectors. The typical materials used for this application are stainless steels (316, 310, and 446) and chromium-plated stainless steels. However, materials with improved corrosion resistance need to be developed for long-term operation (22).

Significant research has focused on the development of alternative materials for both the anode and cathode. Numerous studies (23–25) have examined the corrosion of materials and have served as the foundation for the research being conducted today.

Several metal and alloy materials have been proposed as alternative anodes. The Ni–Cr anodes currently in use yield acceptable performance but are high cost components and are susceptible to mechanical deformation. The Cr in the alloys reacts to form $LiCrO_2$ which aids in stabilizing the porous anode structure in sintering resistance.

However, these materials are susceptible to creep at the high compressive loads encountered in the stack. An anode of Ni–10Cr alloy in a MCFC exhibited a creep of about 4% after a 10,000 h test, which is about four times higher than the goal (18). Metal oxides (such as Al_2O_3, $LiAlO_2$, and ZrO_2) and metals (such as Ti, Co, Cu, and Al) have been incorporated in anode structures (16,18,26–28). Copper-based materials have also been evaluated as a lower cost alternative (26,29,30).

An acceptable cathode material must have good electrical conductivity and structural strength. It must also have a low dissolution rate in molten alkali carbonates to avoid precipitation of the metal in the electrolyte structure. Over 50 materials have been examined to identify suitable materials. Perovskite-type compounds ($Ln_{1-x}M_xM'O_3$ where Ln is a lanthanide element, M is an alkaline earth or transition metal, a M′ is an transition metal) and mixed metal oxides have shown promise. Studies of lithium ferrates have been shown to be a viable candidate for cathodes despite its high solubility (78 ppm Fe) in the Li/K electrolyte at 650°C. In contrast to NiO, this material is independent of the partial pressure of the CO_2 and O_2. In addition, $LiCoO_2$ and $LiCoO_2$-coated NiO have also been shown to have low solubility in the electrolyte. However, it has a very high resistivity (about 300 $\Omega \cdot cm$). Unfortunately, this has a high material cost which may prevent large scale implementation (31–33).

SOLID OXIDE FUEL CELL ELECTRODES

The SOFC technology has emerged from single cell tests in the 1980s to prototype multicell arrays up to 100 kW in the 1990s. These fuels cells can operate at intermediate and high temperatures (650 to 1000°C) and differ from the other fuel cells in that they have a solid ceramic electrolyte. As a result, the electrolyte management problems that are common to the other fuel cells are not an issue. In addition, they do not require CO_2 recycle from the anode to the cathode.

Two basic configurations of SOFCs are being developed: planar and tubular. In addition, the planar has variations such as radial and monolithic. This is shown in Fig. 3. Originally, experimental SOFCs had a planar configuration similar to the PAFCs. This configuration was generally unacceptable due to poor point contact at the three-phase boundaries as well as poor gas edge seals. A tubular configuration was adopted which had bell-shaped cells that overlapped. This also had a three-phase boundary problem as well as problems with thermal expansion coefficients.

Tubular Solid Oxide Fuel Cells. After many iterations, "test tube" configuration was developed. This was partially an outgrowth of the oxygen sensor industry which often used a closed-end tube. These systems operate in the 800 to 1000°C range. 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. In the current configuration, the cathode has a dual purpose: serving as the porous support tube as well as the air electrode. This design allows relatively large single tubular cells to be constructed with the successive active layers deposited with chemical or materials interference with the previously deposited layers. In addition, the manifolding of the oxidant and fuel gases is simplified, and no gas seals are required.

Current designs make use of thin film wall concepts where films of material are deposited one on another and sintered to form the cell structure. The fabrication technique differs for each component. The air electrode provides the inner tube for the cell package. This tube is generally extruded and sintered with one end closed. The anode is generally deposited, to a thickness of about 150 μm, using a slurry process. It is important that these electrodes permit only electronic conduction and interdiffusion of ionic species at operating temperature (1000°C). In addition, they must be resistant to thermal cycling effects. That is, they must have closely matched thermal expansion coefficients and little or no grain growth.

The resistivities for the anode are critical to the performance of the cell. At 1000°C, the anode and cathode should have resistivities of 0.001 $\Omega \cdot cm$ and 0.01 $\Omega \cdot cm$, respectively (34). The electrolyte is the least conductive component in the cell.

Planar Solid Oxide Fuel Cells. Planar cells, on the other hand, generally operate at lower temperatures (650 to 800 °C). This enables the use of metallic interconnects, relaxes some cell construction constraints, reduces internal electrical resistance, and potentially can reduce the cost of materials in the balance of plant. Planar cells require gas-tight seals as part of the manifolding to separate the fuel streams. The seals are a critical technological hurdle for this technology.

Bipolar Configuration. The bipolar configuration is the most common configuration for the planar systems. These systems can either be high or intermediate temperature. This consists of a simple series electrical connection between cells without the need for external cell interconnections. Perpendicular current collection in a cell stack has a lower ohmic polarization than the tubular configuration. However, gas leaks with edge seals and poor triple phase interface contacts generally result in lower performance. Overall, the flat plate design offers improved power density relative to the tubular design. Fabrication and assembly appear to be simpler for the flat plate design. Tape casting, hot pressing, and dry pressing are the most common fabrication methods.

Monolithic Configuration. The monolithic configuration is a more complex bipolar design. The structure resembles the

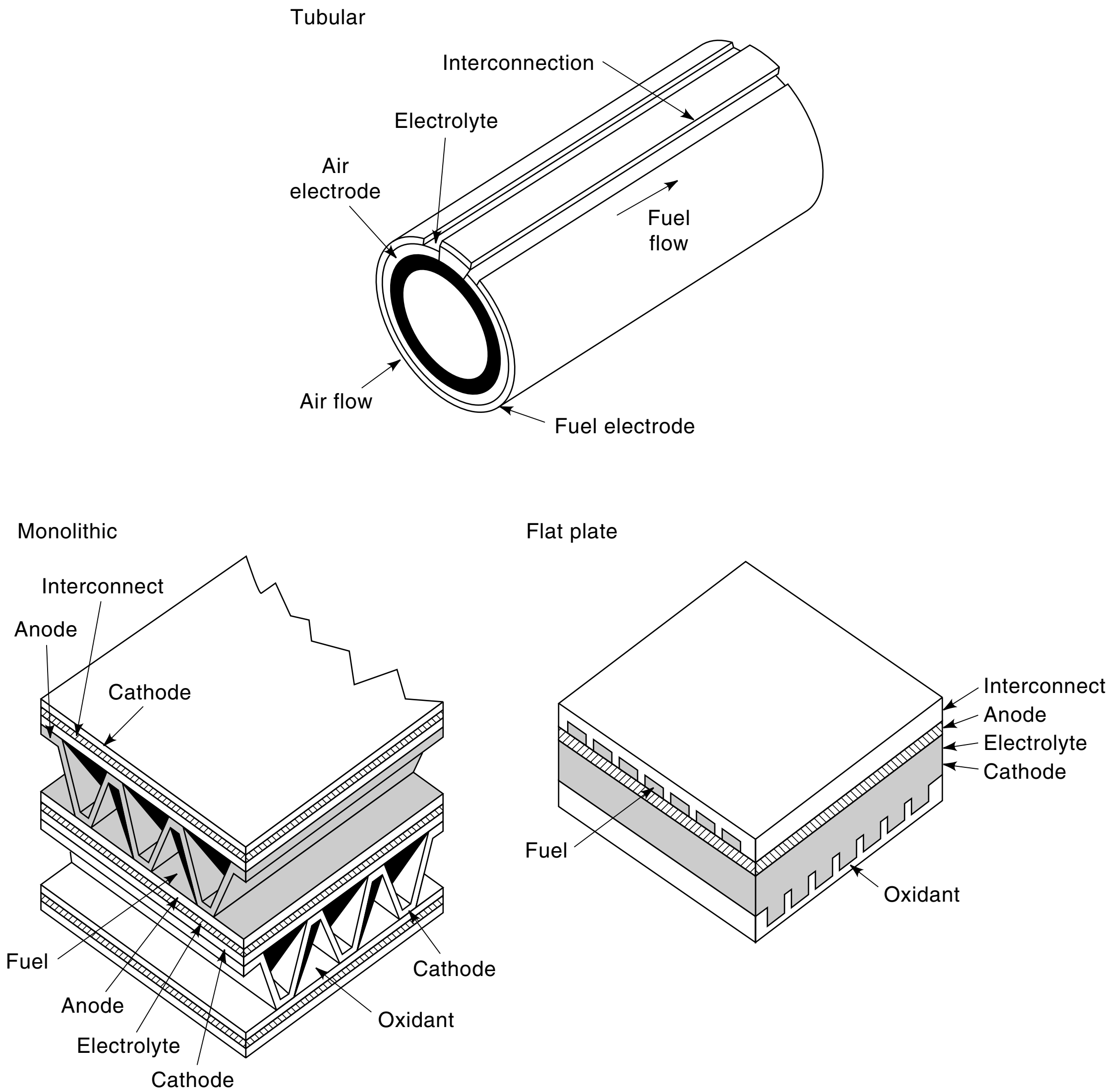

Figure 3. Various solid oxide fuel cell configurations.

corrugated assembly seen in cardboard boxes.The electrodes for this system are taped or calendered and then formed into corrugated structures. These corrugations also serve as the passages for the fuel and air flows. The electrodes are "stacked" with the other components, and the entire "fuel cell stack" is then sintered into a rigid structure. The thermal expansion of the components are critical during heat treating and sintering as well as during stack operation. Residual stresses caused by the differences in thermal expansion coefficients often lead to mechanical failure (35). In addition, it is difficult to cofire metals and oxides in a single atmosphere without either reducing or oxidizing one or the other. The triple-phase boundary of these systems has been shown to have poor contact and show less performance than comparable systems. Lastly, another major challenge is the manifolding for the fuel and oxidant gases against the rigid, uneven, edge surfaces. Calendaring, a variation on tape casting, is the most common fabrication method for the electrodes.

Solid Oxide Component Materials. In general, the materials used for the manufacture of the comonents are similar for both planar and tubular cells. However, the intermediate temperature cells can use different materials, and they are noted separately. The evolution of materials for this technology is shown in Table 5. Much of the current research focuses on the intermediate temperature cells. The materials used in cell components are limited by (1) chemical stability in gaseous (either oxidizing and/or reducing) environments, (2) conductivity, and (3) thermomechanical compatibility. The electrolyte in this fuel cell is a solid, nonporous metal oxide, usually Y_2O_3-stabilized ZrO_2. The cell operates between 650 and 1000°C where ionic conduction by oxygen ions takes place.

Currently, the anode is a $Co{-}ZrO_2$ or $Ni{-}ZrO_2$ cermet containing about 30 mole% Ni or Co. Various modifications to this include partial or complete replacement of the ZrO_2 with CeO_2. Takahashi (36) and Steele (37) have shown that there is enhanced catalytic behavior for the direct oxidation of methane using $Ce_{0.9}Y_{0.1}O_{1.95}$. Unfortunately, they also appear to have poor electronic conductivity and large thermal expansion at higher temperatures. Mixed protonic and ionic conductivity has also shown promise as anode materials (38). In addition, a variety of acceptor doped Perovskite (ABO_3)

Table 5. Evolution of Solid Oxide Fuel Cell Technology for Both Planar and Tubular Configurations

Component	1965	1975	Current
Anode	Porous Pt	Ni–ZrO_2 cermet	30 mole% Ni–ZrO_2 cermet 12.5×10^{-6} cm/cm·°C^a Deposit slurry, dry press, calendar, tape cast
Cathode	Porous Pt	Stabilized ZrO_2 impregnated with praesodymium oxide	10 mole% Sr-doped lanthanum manganite 12×10^{-6} cm/cm·°C^a Deposit slurry/dry press, calendar, tape cast, sinter
Electrolyte	Yttria-stabilized ZrO_2	Yttria-stabilized ZrO_2 EVD	8 mole% yttria-stabilized ZrO_2 10.5×10^{-6} cm/cm·°C^a EVD, dry press, calendar, tape cast
Cell interconnect	Pt	Mn-doped cobalt chromite EVD	10 mole% Mg-doped lanthanum chromite Dry press, calendar, EVD, tape cast, plasma spray
Separator (planar only)			Metal alloy
Support tube (tubular only)	Yttria-stabilized ZrO_2	Yttria-stabilized ZrO_2	None required (air electrode is support for tubular, none for planar)

^{a} Expansion from room temperature to 1,000°C.

can exhibit protonic conductivity. Compositions such as $CaTiO_3$(Fe), $SrZrO_3$(Fe), $LaYO_3$(Sr), and $SrUO_3$ may be acceptable as alternative materials (39).

The cathode is generally an Sr-doped lanthanum manganite ($La_{1-y}Sr_xMnO_3$) containing about 10 mole% Sr. Research has primarily focused on those materials that do not react with the ZrO_2 electrolyte and have good electronic conductivity. Substitution of Co for Mn on the B-site has been attempted, but it appears that only very small amounts of Co can be introduced (40).

Intermediate Temperature Cell Materials. The yttria-stabilized zirconia is a poor conductor if it is operated at temperatures below 800°C. Some improvement is shown if thin film electrolytes are used. Significant alternative materials are being examined for operation at lower temperatures. These systems are simpler, have simpler balance-of-plants, and the potential for lower cost. The most critical component being developed is the electrolyte. The materials for the anode and cathode will need to be compatible with the electrolyte. Some of the low temperature electrolytes being developed are $Ce_{0.8}Gd_{0.2}O_x$, CeO_2-based materials, $(Bi_2O_3)_{0.75}(Y_2O_3)_{0.25}$, $La_{1-x}Bi_xAlO_3$, $Y_{1-x}Ca_xCrO_3$, $(Y_{0.6}Ca_{0.4})_{1-y}Cr_{1-y}O_3$, and salts consisting of Li/K/M, Li/Na/M, and Li/Na/K/M, where M is Ca, Ba, or Sr (41).

Work being conducted on anodes involves alternative materials as well as Cr-stabilized-Ni. Laboratory-scale cells have been successfully testing using Ni-$Ce_{0.8}Gd_{0.2}O_{1.9}$ anodes. Performance improves with thinner components but, in general, is poorer than the Ni-cermet materials (42).

Single-phase porous cathodes appear to have an increased ionic conductivity with high electronic conductivity. Materials that have shown promise include $La_{0.6}Sr_{0.4}Mn_{0.8}Co_{0.2}O_{3-\delta}$ and $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$. Two-phase cathodes, containing both ionic and protonic conductors, perform even better (42).

SUMMARY

One of the main attractive features of fuel cell systems is their expected high fuel-to-electricity efficiency (40 to 70% based on lower heating value of the fuel), which is higher than that of many competing energy conversion systems. In addition, fuel cells operate at a constant temperature, and the heat from the electrochemical reaction is available for cogeneration applications. Since fuel cells operate at near constant efficiency, independent of size, small fuel cells operate nearly as efficiently as large ones. Thus, fuel cell power plants can be configured in a wide range of electrical output, ranging from watts to megawatts. Fuel cells are quiet and operate with virtually no gaseous or solid emissions. The two major impediments to the widespread use of fuel cells are: (1) high initial cost and (2) endurance operation; it is these two aspects which are the major focus of technological effort.

The various fuel cell types (AFC, PEFC, PAFC, MCFC, and SOFC) have significantly different operating temperatures. As a result, their materials of construction, fabrication techniques, and system requirements differ. These distinctions result in individual advantages and disadvantages which govern the potential of the various cells to be used for different applications.

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ELECTROCHEMICAL SENSORS. See Intelligent biosensors.

ELECTRODES, ELECTROCHEMICAL. See ELECTROCHEMICAL ELECTRODES.