Fuel Cell Technology**

Samuel Stucki*

Since fossil fuels are likely to remain the main energy supply in the near future, conservative energy use and the application of highly efficient energy conversion processes are still the only way to alleviate the CO_2-problem. It has long been recognized that «cold burning» of fuels in an electrochemical «fuel cell» results in an energy conversion with potentially very high efficiency. Fuel cells were first put to work with pure hydrogen as the fuel in space technology in the 1960's. The principles of the technology were developed during that time. The subsequent developments have focussed on terrestrial applications and on systems compatible with the use of fossil fuels. There is a large number of possible combinations of electrode and electrolyte materials to build a fuel cell. The five major types are discussed in the present article. The hitherto most advanced system is the phosphoric acid fuel cell (PAFC), which can be operated on reformed natural gas as the fuel; PAFC technology has been demonstrated on a 4.5 MW scale. More advanced fuel cell systems operating at temperatures above 600° C are expected to become an efficient and clean alternative to the established thermal processes in the production of electricity from coal. Fuel cell types operating at low temperatures are expected to play a future role in decentralized energy use, transportation, and «hydrogen technology».

Samuel Stucki: Born 1944 in Ins (BE). Diploma in chemistry from Universität Bern, PhD thesis in the field of electrochemistry from 1970 to 1973 at the Institut für Anorganische, Analytische und Physikalische Chemie with Prof. E. Schmidt. From 1973 to 1976 he worked at the Fritz-Haber-Institut in Berlin as a postdoctoral fellow with Prof. H. Gerischer. Since 1976 he has been working at the Brown Boveri Research Center in Baden, where he established an electrochemical research group. The main activities of his group have been in the fundamentals and the development of an advanced water electrolysis technology for hydrogen generation, electrocatalysis, new electrochemical processes, and more recently membrane separation processes.

1. Introduction

It has long been recognized that «cold burning» of fuels in an electrochemical cell results in an energy conversion with potentially very high efficiency. So-called fuel cells were defined by *Wilhelm Ostwald* at the end of the nineteenth century: a fuel cell is an electrochemical cell, which transforms the chemical energy of a fuel and an oxidant into electric energy, whereby the electrode processes take place in an invariant system of electrodes and electrolyte[1]. The second part of this definition distinguishes fuel cells from primary and secondary batteries, where the electrode materials are usually identical with the fuel or the oxidant. Numerous attempts to apply the theoretical ideas to a practical cell have been made since the turn of the century. Most of the early attempts failed because of materials problems, which could not be mastered at that time, or because they were too ambitious in that they were trying to use «natural» fuels such as coal, gas, or petroleum directly.

The foundations of modern fuel cell technology were laid technically and scientifically in the 1950's: pioneering work on fuel cell technology was done by *Bacon*[2], the fundamentals of electrochemical kinetics and electrocatalysis were established at the same time. The alkaline cell of *Bacon* was the first design to use porous gas electrodes for the synthetic fuel hydrogen and the oxidant (oxygen or air). The nickel electrodes, which were used as anodes and cathodes, had fine pores on the electrolyte side and coarse pores on the gas side. In this way it was possible, in a pressurized system, to establish the three-phase-boundary gas/liquid/solid which is important for the attainment of high current densities (see Section 2.3).

* *Correspondence:* Dr. S. Stucki
Asea Brown Boveri Forschungszentrum
CH–5405 Baden

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Bacon's cell served as a model for the fuel cell system selected by the American space administration (NASA) for their Apollo and later for their Space Shuttle programs. The American space program, in fact, brought about the first technical application of fuel cells. Cryogenically stored hydrogen and oxygen were used as reactants. The solid polymer electrolyte fuel cell (SPEFC), which was developed by General Electric, was used for power generation and drinking water production for the Gemini missions[3]. As its name implies, the solid polymer electrolyte fuel cell consists of a solid electrolyte, contacted by porous electrode structures. The solid electrolytes or rather, ion-exchange membrane materials available at that time were only of very limited stability. This was the main reason why they were ruled out for the subsequent Apollo program, which involved longer flights. Ion-exchange membrane cells were reinvestigated later, after a new class of highly chemically resistant membrane polymers became available.

The development work on alkaline cells, which had been started by a large number of competing companies, was largely given up after the successful landing on the moon. Some activity continued or was taken up later, especially in Europe (Siemens, ELENCO)[4]. In the USA, in an attempt to bring space technology down to earth, emphasis was shifted away from alkaline cells to the phosphoric acid cells and to the high temperature cells, which until today have remained the systems receiving most attention and funding. They will be discussed in more detail below. The reason for ruling out alkaline cell space technology for terrestrial applications is the inherent incompatibility of the alkaline electrolyte with the carbon dioxide present in any fossil-derived fuel and in the oxidant air. Although CO_2-scrubbing from steam-reformed natural gas (a mixture of H_2 and CO_2) can be done efficiently, the decision of the American funding agencies (DOE mainly) was to concentrate on systems, which would be compatible with CO_2. Phosphoric acid fuel cells (PAFC) are today the most advanced systems for civilian energy conversion applications. PAFC systems of some ten megawatt per unit are expected to contribute to the electric utility market in the years to come, especially for local power generation in densely populated areas with restrictive immission regulations.

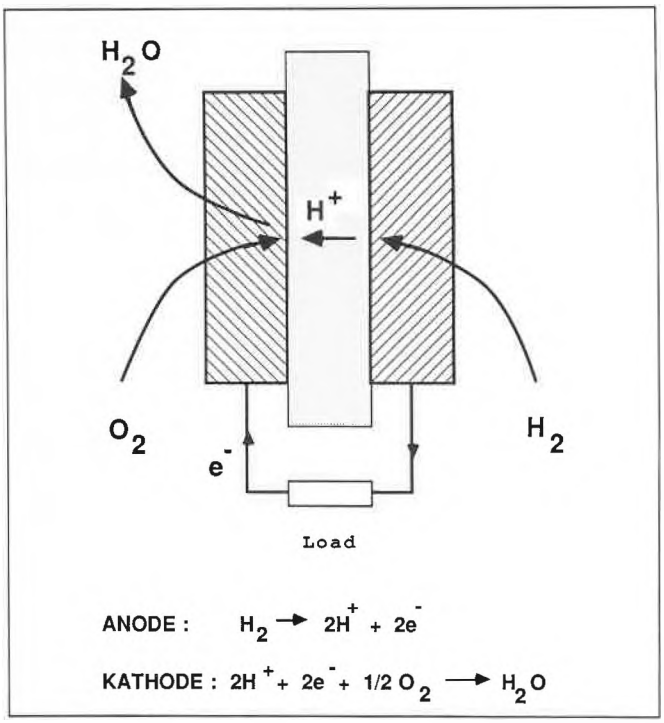

Fig. 1. Schematic representation of processes taking place in a fuel cell with acid electrolyte. Hydrogen gas is oxidized at the anode/electrolyte interface to hydrogen ions. The hydrogen ions are transported in the electrolyte to the cathode, where the reduction of oxygen gas to water completes the cell reaction. The processes build up an electric potential between the electrodes and electric current can be drawn, if the electrodes are connected via a load resistance.

2. Fundamentals of Fuel Cell Reactions

2.1. Cell Characteristics

Fig. 1 shows the electrochemical processes taking place in a fuel cell with an acid electrolyte. Hydrogen gas is fed to the anode/electrolyte interface via the pores of the anode current collector structure. At the interface, hydrogen is oxidized and the hydrogen ions formed are transported by the electrolyte to the cathode, where molecular oxygen is reduced to water. The reaction product water is usually removed from the electrolyte by evaporation. The electric voltage, which the cell can produce optimally, corresponds to the free enthalpy change of the hydrogen combustion reaction:

$$U_{max} = -\Delta G/nF$$

where ΔG: free enthalpy change, n: number of electrons transferred by reaction equivalent, F: Faraday constant. If the cell is set up to produce finite currents, the cell voltage decreases due to irreversibilities in connection with the electrode reaction and electric conduction. This is visualized in Fig. 2: the current-voltage relationship of a fuel cell can be divided into basically three sections.

Section I: Losses due to irreversibility of the electrode reactions; the current depends exponentially on overvoltage $(U-U_{max})$. In hydrogen/oxygen cells the irreversibility can be localized at the cathode (oxygen electrode).

Section II: Linear region; the slope of the curve is mainly given by the ohmic resistance of the cell, i.e. by the electrolyte resistance.

Section III: Limiting current region. If the cell is operated using mixtures of reactant gases with inert gases (e.g. air as an oxidant or H_2/CO_2 as fuel), the current may become limited by mass transfer to the electrodes.

Technical cells are operated with optimum power density in the linear part of the curve.

2.2. Temperature of Operation and Efficiency

The maximum efficiency of a fuel cell is usually related to the heating value of the fuel:

$$\eta_{max} = \frac{\Delta G}{\Delta H} = 1 - \frac{T\Delta S}{\Delta H}$$

where ΔH: enthalpy change, ΔS: entropy change, T: temperature. For hydrogen/oxygen cells, the quotient $T\Delta S/\Delta H$ is positive, i.e. the maximum attainable efficiency will decrease linearly (assuming ΔS and ΔH to be temperature independent) with increasing temperature from 84% at room temperature to 54% at 1000 °C. In spite of this, ongoing development trends are aiming at higher temperatures: 200 °C (PAFC), 600 °C (cells with molten alkali metal carbonates as electrolyte: MCFC), or even up to 900–1000 °C (cells with ceramic solid electrolytes: solid oxide fuel cells, SOFC). The reasons for raising the temperature, are:

a) Ohmic losses and electrode irreversibilities decrease with increasing temperature.
b) Electrode surfaces tend to be less prone to poisoning at elevated temperatures.

c) Other fuels besides hydrogen can be processed in the cells at high temperatures (SOFC and to a certain extent MCFC).
d) If the cells are operated at high temperature, the waste heat can be utilized in a subsequent thermal process (e.g. a steam turbine).

General disadvantages of raising the operating temperature of a fuel cell are increased materials problems (corrosion, embrittlement etc.) and the loss of cold start-up capability. In terms of electric efficiency, the thermodynamic losses at elevated operating temperatures are roughly compensated for by the more reversible kinetics. Typical efficiencies, which can be reached by fuel cells of all types at economically feasible current densities, are in the order of 40 to 60% (related to the heating value of hydrogen).

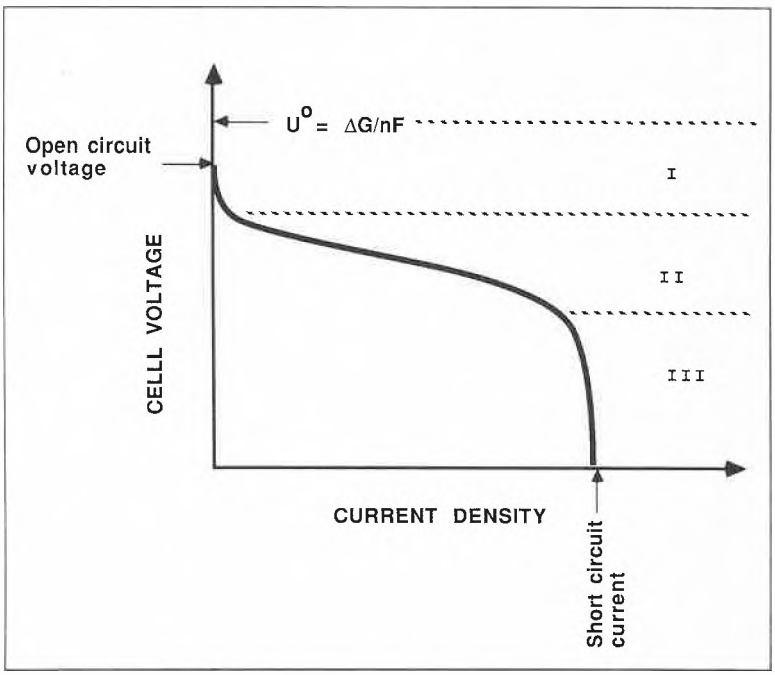

Fig. 2. Current/voltage characteristic of a fuel cell. The curve can be divided into three domains (dotted lines) according to the relative predominance of the processes limiting the efficiency of the cell with increasing current density: (I) At low current densities, the efficiency (i.e. the cell voltage) is limited by irreversibilities of the electrode reactions. (II) At intermediate current densities, the curve approaches a straight line, in this range the efficiency is limited by the ohmic resistance of the cell. (III) If the fuel cell is operated with mixtures of the electroactive and inert gases (e.g. air as oxidant and/or reformate as fuel), a limiting current is observed due to mass transfer limitations.

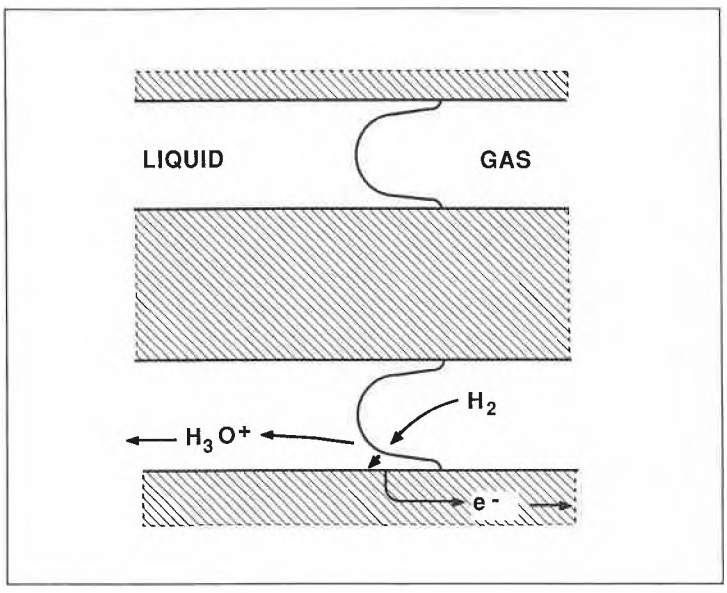

Fig. 3. Schematic cross section of a pore of a gas electrode. The reactive gas (formulated for hydrogen) reaches the reaction site by diffusing through a thin film of electrolyte which is wetting the walls of the pore.

2.3. Electrodes and Electrolytes for Fuel Cells

The main technological problem with fuel cell electrodes has been to provide fast transport of the reactant gases to the electrolyte/electrode interface. Under normal conditions, the gases are only slightly soluble in aqueous electrolytes (millimoles per liter). The transport-limited current would be impractically low if cells were operated in gas-saturated liquid electrolytes. The problem has been solved by using porous electrode structures exhibiting a so-called three phase boundary, which is characterized by short diffusion paths in the condensed phase (the diffusion path has to be much shorter than the diffusion layer at a solid surface in contact with a stirred electrolyte). Fig. 3 sketches the three phase boundary in a pore of a fuel cell electrode. Mass transfer from the gas phase to the electrode is believed to proceed mainly via the wetting meniscus where diffusion should be fastest. In an optimum electrode design, the reaction zone, i.e. the transition zone from gas to electrolyte, is engineered so that it coincides with the zone of highest catalytic activity, which normally means the zone with the highest concentration of noble metals. In order to control the position of the reaction zone in an electrode, different approaches are possible:

a) For liquid electrolytes: By providing a dual porosity of wide and narrow pores, the liquid and gas phase will be distributed according to capillary forces, which strongly depend on pore diameter. A similar effect can be achieved by changing the wetting properties of the materials constituting the pore surfaces. The latter technique is applied in alkaline and phosphoric acid cells using poly(tetrafluoroethylene) (PTFE) to provide water repellant pores.
b) For solid electrolytes, the three phase boundary is fixed and defined by the geometry of the contacts between the electrode and the solid electrolyte.

The advantage of the liquid electrolytes is that the reaction zone usually seems to be more extended than in solid electrolyte designs. The disadvantage is that the reaction zone may migrate within the electrode during prolonged operation with the danger of the electrode either drying out or drowning. As a matter of fact, electrode wetting and drowning is one of the major phenomena limiting the useful life of cells with liquid electrolytes. Fuel cells with solid electrolytes (SPEFC and SOFC) are known to exhibit the best long-term performance of all systems.

2.4. Electrocatalysis

Electrocatalysis plays an important role in improving the performance of fuel cells. Both electrode reactions involve more than one electron transfer per molecule of reactant and hence involve adsorbed intermediates, the energy of which can be influenced by catalysis. Much fundamental work has been performed on the electrocatalysis of the hydrogen oxidation and the oxygen reduction reactions. The latter is technically of greater relevance because the oxygen electrode is known to contribute most of the polarization losses of low- to medium temperature cells and hence has more potential for improvement. It is beyond the scope of this review to cover the abundant literature on fuel cell electrocatalysis. The result of all these efforts at the present time seems to be that one still cannot do without noble metal catalysts for technically practicable cells. Considerable progress has, however, been made in terms of catalyst utilization (i.e. specific catalyst loadings) and advanced, bimetallic, noble metal based catalysts[5].

3. Types of Fuel Cells

A large number of potential configurations of electrodes, electrolytes, and systems have been proposed in the literature. The systems that are being investigated and/or developed to a full scale are listed in Table 1. Emphasis is put on fuel cell systems with a potential for applications in the range from some ten kW to some MW. Excluded from this list are the direct methanol fuel cells (DMFC), which have been paid more attention recently, but which, according to most experts, will not play an important role in medium- to large scale energy conversion due to electrode irreversibilities at the fuel electrode[6].

3.1. Alkaline Fuel Cells (AFC)

At moderate temperatures (below 200° C), the overvoltage for the oxygen reduction reaction using standard electrocatalysts is smaller in alkaline than in acidic media. Alkaline cells therefore achieve the highest efficiencies of all types of fuel cells, which makes them attractive for space applications. The most advanced cells have been optimized for extremely high power density and high efficiency: cell voltages of 0.8 V at 5A/cm^2 have been achieved with cells operating at 150 °C and a pressure of 10 bar. These data correspond to a power density of 40 kW/m^2 with an efficiency of 55%. These cells have not, however, been optimized with respect to cost: only expensive materials withstand the operating conditions of such cells. Low-cost AFC's for terrestrial applications have been developed by various laboratories. Most of the work on alkaline cells was performed in the 1960's and has been discontinued since. The Belgian-Dutch consortium ELENCO are about to demonstrate AFC technology for transportation (buses)[7].

Alkaline cells rely on pure hydrogen as a fuel. The availability and cost of clean hydrogen will decide whether AFC will play an important role in the future. A Swedish study has come to the conclusion that the removal of CO_2 and catalyst poisons from a coal gas could be done competitively in order to operate low-cost AFC's using coal as a primary fuel. Certainly AFC technology could become important in a «Hydrogen Energy System», where the major source of hydrogen would be water electrolysis[4].

3.2. Ion-Exchange Membrane Fuel Cells (IEMFC)

The solid polymer electrolyte cell (SPEFC) was the first cell which was used in a space mission. The cell consists of an ion-exchange membrane, contacted on both sides by catalyst layers acting as the gas electrodes. The key element of the technology is the ion-exchange membrane. The first cell had a limited lifetime because the membrane materials (sulfonated polystyrene) did not withstand the reactive intermediates present at the surface of the electrocatalysts during operation of the cells. A breakthrough in membrane technology was the development of highly resistant perfluorinated cation-exchange membranes (Nafion, DuPont) which led to new applications of the basic membrane cell design in processes like chlor-alkali electrolysis and water electrolysis[8]. Although the membrane fuel cells using Nafion membranes are considered to be the most reliable technology in terms of long-term stability, exhibiting at the same time good efficiencies at quite high power densities, the system has not been developed for purposes other than military. The reasons for this are the high costs of membranes and high catalyst loadings in the classical design of the cell[5]. Recent progress in reducing catalyst loadings of membrane cells by a factor of almost 10 and the development of a new membrane by Dow[9], with much improved performance in terms of power density of the cell, call for a re-evaluation of the membrane fuel cell for non-military applications. Using the new membrane from Dow Chemical, the Canadian development group BTC[9] have been able to increase the power density of the cell by a factor of 4. This means a drastic reduction of specific cell costs.

Membrane cells have been discussed for regenerative applications, i.e. they can basically be operated in both the fuel cell mode and in the electrolyzer mode, provided that suitable electrocatalysts can be found. This possibility would open up the use of such a system for energy storage. Energy storage efficiencies of up to 50% should be possible. This application is still far away from being achieved.

The most promising applications of the membrane fuel cell are in the area of small-scale (a few hundred kW) decentralized electricity and heat co-generation for buildings and possibly for transportation.

3.3. Phosphoric Acid Fuel Cells (PAFC)[10]

Phosphoric acid fuel cells have been developed in USA by United Technologies, Engelhardt, and Westinghouse since the early 1970's. Japanese developers started ambitious programs later, with the goal of commercializing the technology within the next decade.

The main components of the cell are made from carbon (i.e. the electrode structures and the bipolar separator plates separating individual cells in a bipolar cell stack). The electrolyte is phosphoric acid with a few percent of water dissolved in it. The electrolyte is immobilized in a porous inert matrix made of a silicon-based material. Cells have been scaled up to an electrode area of 10 ft^2 ($\approx$ 1 m^2). An extensive demonstration program has been initiated: a 4.5 MW demonstration plant was successfully built by UTC and operated in Tokyo; a 11 MW demonstration plant is planned for the immediate future. A number of plants in the power range from 40 to 200 kW has been operated at various sites (co-generation of heat and electricity in large buildings). The fuel considered for use in phosphoric acid cells has been natural gas exclusively.

A typical PAFC power plant consists of a steam reformer, the fuel cell stack, and a DC/AC-converter (see Fig. 4). The steam reformer, which produces a hydrogen/carbon dioxide mixture from methane, operates with steam from the fuel cell. The steam reforming process runs at a temperature of ca. 500 °C. The extra heat required

Table 1. Types of fuel cells.

Type	Electrolyte	Temperature [°C]	Electrodes	Fuel
Alkaline (AFC)	NaOH, KOH – circulating – matrix	80–150	C, Pt, Ag, Pt/Au	H_2 (CO_2-free)
Membrane (IEMFC,SPEFC)	Ion-exchange membrane, aqueous	80–100	C/Pt	H_2
Phosphoric acid (PAFC)	H_3PO_4, matrix	$\approx$ 200	C/Pt	H_2, CO traces
Molten Carbonate (MCFC)	Li_2/K_2CO_3, matrix	650	Ni, NiO	H_2, CO, (CH_4)
Solid Oxide (SOFC)	ZrO_2-ceramic	800–1000	Ni, cermet, oxide	H_2, CO, CH_4

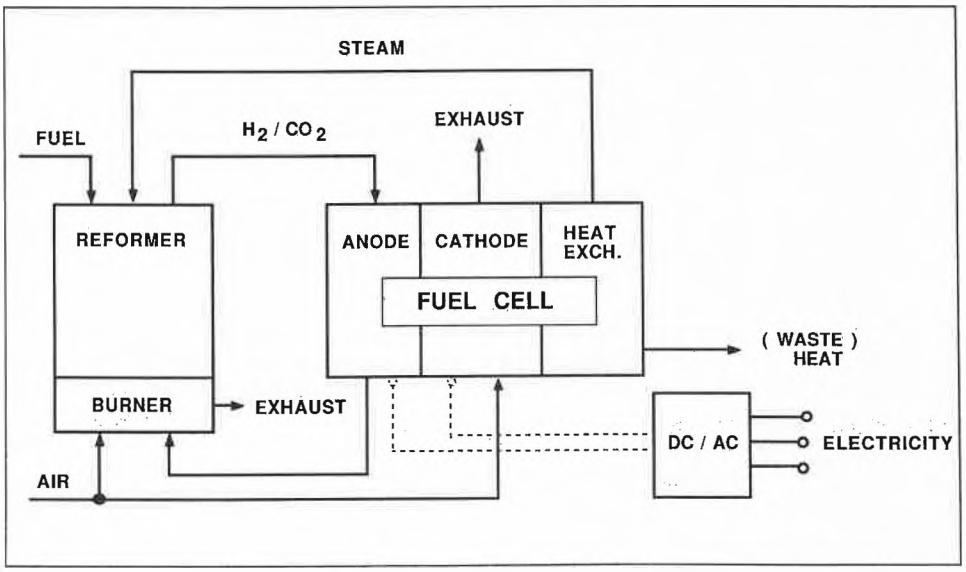

Fig. 4. Simplified flow-sheet of a fuel cell (PAFC) power plant operating with natural gas as a fuel. The energy input required by the fuel processing unit (steam reformer) is provided by the fuel cell: the off-gas from the fuel electrode, which is not completely depleted of hydrogen, is burned to provide the necessary temperature of the reformer. Steam is produced from the fuel cells cooling loop.

by the process is provided by burning the excess hydrogen from the anode exhaust of the fuel cell. The overall efficiency of a PAFC power plant, related to the heating value of the methane, has been demonstrated to be in the order of 42%.

3.4. Molten Carbonate Fuel Cells (MCFC) [10]

As with most of the presently investigated fuel cell systems, the roots of the MCFC go back to the 1950's. The first work on that system was performed in Holland in *Ketelaar*'s laboratory[11]. The MCFC has been taken up since by a number of developers in USA and Japan; more recently, European programs have been initiated.

The MCFC operates at 600 °C and uses, as its name implies, a K_2/Li_2CO_3-eutectic as the electrolyte. The chemistry of the MCFC is somewhat different from the other fuel cell systems: the electric current transports a stoichiometric amount of CO_2 through the electrolyte from the cathode to the anode according to the reaction scheme

$$0.5\ O_2 + CO_2 + 2e^{\ominus} \rightarrow CO_3^{2\ominus} \text{ (cathode)}$$
$$CO_3^{2\ominus} + H_2 \rightarrow H_2O + CO_2 + 2e^{\ominus} \text{ (anode)}$$

This implies that the cell has to be operated with recycling of carbon dioxide, which is available from the fuel processing unit. It is clear that the MCFC is designed for use with fossil fuels, mainly coal gas. It is compatible with CO, unlike the PAFC, which can easily be poisoned by CO levels above a few ppm. Recent developments are also focussing on the incorporation of reforming catalysts into the cell stack, which will ultimately lead to a system capable of being operated directly with natural gas.

The development of MCFC technology still requires extensive basic research, mainly connected with materials problems. The carbonate melt is very corrosive and improved materials for both the electrodes and the electrolyte matrix are required. MCFC technology is not expected to be available on a commercial scale before the turn of the century. Clearly the development of MCFC technology aims at larger power plants, where its main advantages, i.e. the possibility of utilizing coal and the use of the relatively high temperature level of the exhaust in a steam turbine, can be exploited.

3.5. Solid Oxide Fuel Cells (SOFC) [10]

Yttria-stabilized zirconia exhibits ionic conductivity for oxygen ions at temperatures above 800 °C, which is sufficient for it to be applied as an electrolyte in a high temperature fuel cell. Using high temperatures brings several advantages, such as reduced restrictions on the choice of fuels and utilization of waste heat (see Section 2.2). The combination of these advantages with the good stability of a cell design using a solid electrolyte makes this system look very atrractive for power plant applications.

Electrolyte and electrodes of a SOFC consist of ceramic materials: zirconia is the main constituent of the oxygen ion conducting electrolyte, the cathode is usually a conducting oxide with perovskite structure, the anode is made from a Ni cermet. Modern designs of the cell apply the electrode/electrolyte composite structure layer as thin ceramic films on top of a porous substrate. Fig. 5 shows the tubular design which is employed by the now leading developer of SOFC technology (Westinghouse). Bundles of such tubes are connected to a SOFC battery in the pilot design. Many problems remain to be solved in SOFC technology. They have to do

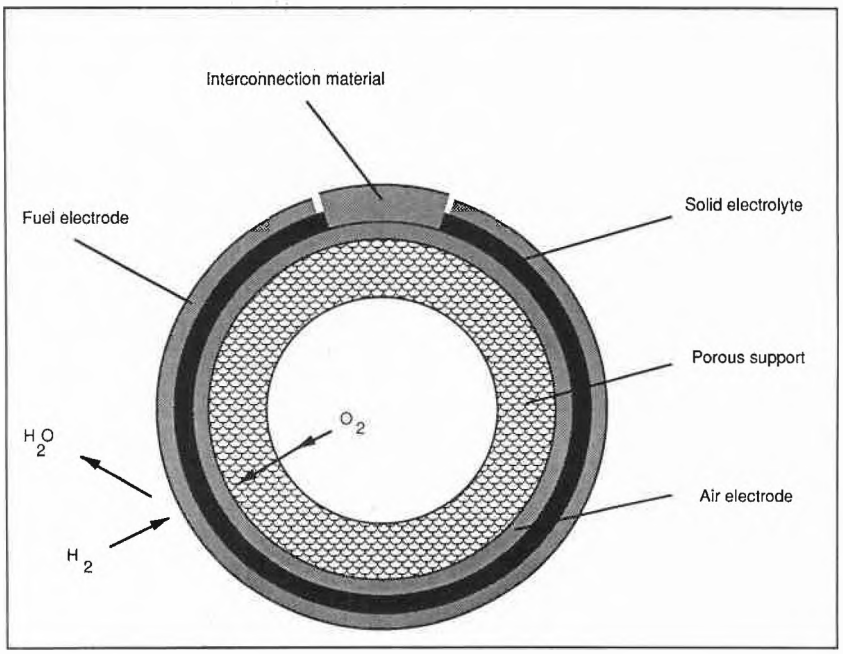

Fig. 5. Cross section of a solid oxide fuel cell (SOFC). The electrodes and the electrolyte are formed by successive thin ceramic layers on a porous carrier tube (Westinghouse design). The electric contact to the inner electrode is made by an electronically conducting interconnecting material.

mainly with the scale-up of the modules, the stability and conductivity of interconnecting materials (i.e. the electronically conducting leads from cell to cell and eventually to the electric bus bar at ambient temperature), and with advanced ceramic processing technologies to ensure high reliability of the mechanically delicate structures.

If successful, the SOFC technology seems to be the most promising technology in advanced power generation from fossil resources. Efficiencies up to 65% for combined cycle power plants with coal gasification, SOFC, gas turbine, and steam turbine as consecutive steps have been projected. Solid oxide cells can also be operated in the electrolysis mode, splitting steam into oxygen and hydrogen, without major changes in the cell design.

4. Economic Considerations and Conclusions

The previous sections have shown that fuel cell technology, which allows efficient and environmentally safe conversion of fossil fuels into electric energy, is already available. It has a potential for further improvement in the future with the implementation of the more advanced technologies under investigation. The commercialization of fuel cells on a significant scale will depend on their economic competitiveness with the established thermal energy conversion processes.

The cost of producing 1 kWh of electric energy depends on the cost of the primary fuel necessary to produce this energy and on the investment cost of the energy conversion device. Since fuel cells promise better efficiencies, their competitiveness will increase with increasing cost of fossil fuels. The decisive cost factor however is the specific investment cost. The specific cost of PAFC systems at present is in the order of 5500 $/kW of installed capacity[12]. This figure is far too high to be competitive with gas turbine technology. An important fraction of the total cost is the cost of the cell stack. Due to the modular design of fuel cells, the specific stack cost is not dependent on the scale of the system, contrary to thermal machines, which show a considerable cost regression with scale. The cell stack is, however, the part of a fuel cell plant which offers most potential for cost reductions. The individual cells of any one of the techniques discussed above lend themselves to mass production with consequent cost reductions (as an example: a 10 MW PAFC plant will incorporate some 7000 identical cells of 1 m^2 each). The projected cost of PAFC technology will reduce to 1000 $/kW or less with mass production. Of course, mass production can only be introduced if a demand for large quantities of fuel cells already exists. This will only be the case if fuel cells are sufficiently economic – a chicken-and-egg dilemma! In any case, it will cost a large sum to subsidize the technology until its costs have come down enough to be competitive. Estimates of the sums required for PAFC technology to break even are in the order of 500 Mio $ over the next 15 years[13].

Those cell costs, which cannot be reduced by mass production, are mainly the material costs of the cell components. Here, further research and development will be needed which focuses on reducing the quantity of expensive materials (in particular noble metals) and on increasing the specific power output of the cells (kW per m^2 or better per kg of cell material). The latter objective can be attained by optimizing the electrolyte with respect to conductivity. The example of the membrane fuel cell (Section 3.2) shows that an improvement in the electrolyte can bring about a marked improvement in the process economy[9].

The costs of the peripheral equipment of a fuel cell power plant (fuel reformer, electric power conditioner, etc.) are more or less accessible to reliable costing, since these parts represent established technology. The fuel processing unit is one of the components that conventional thermal engines can do without and which therefore adds to the bill of a fuel cell system. On the other hand, a fuel cell plant using decontaminated hydrogen as a fuel will not need any flue gas treatment. Again, it seems that more stringent ecological legislation will favor the fuel cell.

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