Review

ADVANCED ELECTROCHEMICAL ENERGY SYSTEM - AN OVERVIEW ON FUEL CELLS

S. Gupta and J.D. Pandya*

Tata Energy Research Institute, 90, Jor Bagh, New Delhi 110 003

INTRODUCTION

Most of our useful energy is obtained from the conventional conversion processes:

Chemical energy ⟶ Heat ⟶ mechanical energy ⟶ electrical energy

These conversion processes transform energy stored within the chemical bonds of hydrocarbon fuels to heat energy after combustion. The maximum amount of useful power that can be obtained from this energy is governed by the principle of a Carnot engine. A fuel cell is an electrochemical device in which the chemical energy of a conventional or non-conventional fuel is directly and efficiently converted into low voltage, direct current electric energy. This power output can be multiplied by connecting a number of cells in series or parallel arrangements to get the desired type of cell. Unlike other batteries, the fuel cell does not consume its electrodes in this process but instead uses up fuel and oxidant, fed continuously from outside the cell. One of the major advantage of such a device is that, the Carnot limitation on efficiency does not apply, since the electrochemical oxidation can be carried out isothermally. Moreover, the fuel cell power plant, consisting of fuel conditioner, fuel cell subsystem, and power conditioner, eliminates the high temperature combustion and the losses associated with mechanical-to-electrical conversion processes found in all energy conversion systems that produce electrical power. The efficiency of a fuel cell power plant can, therefore, be much higher than any conventional power plant. A coal fired thermal power plant operating under optimal conditions can yield a 35 percent efficiency. A nuclear power station can at best attain an efficiency of 30 percent. On the other hand, a fuel cell not only generates electricity at 40 percent efficiency, but its end products (hot water and steam) can also be harnessed to generate additional power. Thus the overall efficiency of the fuel cell power plant adds up to between 70 and 80 percent. Lastly, unlike other power plants, it can use a variety of fuels like hydrogen, methanol, ethanol, natural gas, fuel oil, biogas etc. This paper describes the state of the art of fuel cell technology. The basic principles of this technology have been dealt with extensively. Various types of fuel cells that have been developed over the years have also been discussed and classified. A seperate section summarizes the current status of this technology. Lastly, the paper discusses potential applications in India.

BASIC PRINCIPLES

A fuel cell operates electrochemically or more literally "chemico-electrically" as shown in Figure 1. This cell is actually a reactor, wherein hydrogen as the feed stream or fuel is conducted into the empty space paralleling the porus, conducting anode. This anode can be made of porous carbon with a metal catalyst like platinum, which chemically changes the hydrogen atoms to positively charged hydrogen ions and electrons.

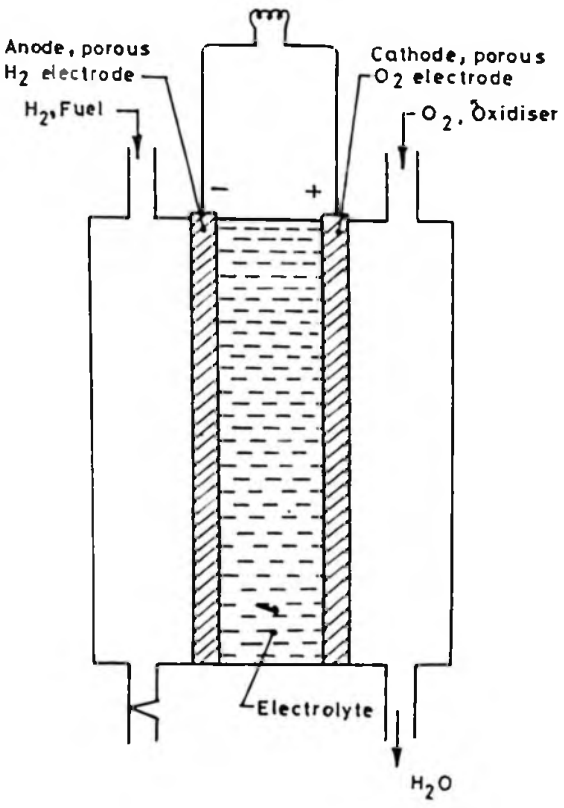


Fig. 1 Schematic of H_2-O_2 Fuel Cell.

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$$H_{2(g)} \xrightarrow{\text{anode}} 2H^+ + 2e^- \quad \ldots\ldots (1)$$

The electrons leave the anode, perform electrical work and enter the cathode. Meanwhile, the positively charged hydrogen ions migrate through the electrolyte attracted by the electronegative oxygen from the cathode.

$$2H^+ + 1/2\ O_{2(g)} + 2e^- \xrightarrow{\text{Cathode}} H_2O + \text{Heat} \quad ..(2)$$

To complete the reaction, the oxygen pulls in the electrons from the outer circuit, thus water is generated and discharged from the cell. The overall fuel cell reaction is obtained by combining equations (1) and (2).

$$H_{2(g)} + 1/2\ O_{2(g)} \longrightarrow H_2O_{(g)} \quad \ldots (3)$$

The electrical work done by a fuel cell (W_{el}) is given by the amount of charge that flows from the cell multiplied by the potential difference of the cell E, the driving force that causes them to flow. The amount of charge is given by the product of the number of moles of electrons involved in the cell reaction times the number of coulombs per mole of electrons. Thus,

$$W_{el} = nFE \quad \ldots (4)$$

where n represents the number of moles of electrons transferred from anode to cathode during one act of the overall reaction, F is the Faraday constant. For a steady gas flow fuel cell system in both temperature and pressure equilibrium, it can be shown that the maximum useful work is equal to the decrease in Gibbs free energy, i.e.,

$$W_{max.} = W_{el} = -\Delta G \quad \ldots (5)$$

combining equations (4) and (5)

$$\Delta G = -nFE \quad \ldots (6)$$

The potential difference E in equation (6) is the reversible potential of the cell, that is the potential measured under circuit conditions. The equilibrium cell potential E is related to the activities of the species involved in the reaction by the Nernst equation [1].

$$E = E^\circ + \frac{RT}{nF} \ln \frac{\text{(Product of the activity of reactant species)}}{\text{(Product of the activity of product species)}} \quad (7)$$

Where E° is the standard electromotive force or the standard cell potential when all the species involved are at unit activity. E° is related to the standard free energies of the species by

$$\Delta G^\circ = nFE^\circ \quad \ldots (8)$$

The thermodynamic efficiency of energy conversion in a fuel cell can be defined as the ratio of electrical energy produced to the total thermal energy that can be released in direct chemical reaction. At constant temperature, the change in Gibbs free energy is given by:

$$\Delta G = \Delta H - T\Delta S \quad \ldots (9)$$

Where ΔH is the enthalpy change for the fuel cell reaction, often called the heat of reaction or heat of combustion. This enthalpy change is the maximum amount of thermal energy that can be produced isothermally, when fuel is burnt in the conventional manner. ΔS is the entropy change for the process and for the reversible process the quantity $T\Delta S$ represents the isothermal heat transfer. Thus Gibbs free energy change, as per equation (5), is the maximum amount of chemical energy that can be converted into electricity. The fuel cell involves exothermic reaction and thus rejects heat. From the second law of thermodynamics, heat rejection results in a decrease of entropy. For most chemical reactions of interest both ΔG and ΔH are normally negative and thus the decrease in entropy associated with a heat rejection causes ΔG to be numerically smaller than ΔH. The quotient $\Delta G/\Delta H$, therefore, provides a measure of the thermodynamic efficiency of the fuel cell [2].

$$\eta_T = \frac{\Delta G}{\Delta H} = 1 - \frac{T\Delta S}{\Delta H} \quad \ldots . (10)$$

Equation (10) determines the thermodynamic limits to which the heat of combustion can be recovered as electric energy. The ideal efficiency of a fuel cell is always less than unity, even in a cell operating reversibly, so long as heat ($T\Delta S$) is being rejected. Table 1 lists the thermodynamic characteristics of important fuel cell reactions. It is interesting to observe the changes in the thermodynamic efficiency of a simple hydrogen-oxygen fuel cell, as its operating temperature is raised from 298°K to 2000°K. Table 2 lists the functions ΔH° and ΔG°, the standard heat of reaction and the change in Gibbs free energy respectively for the reaction (3). The

TABLE 1

THERMODYNAMIC CHARACTERISTICS OF IMPORTANT FUEL CELL REACTIONS [2]

Reaction	T (°K)	ΔG (KJ/mol)	ΔH (KJ/mol) Ideal (%)	Efficiency Ideal (%)
$H_2 + (1/2)\,O_2 \longrightarrow H_2O\,(L)$	298	-237	-285	83.15
$\longrightarrow H_2O\,(G)$	298	-229	-242	94.6
$\longrightarrow H_2O\,(G)$	773	-203.5	-246	82.7
$(2/3)NH_3+(1/2)O_2 \longrightarrow (1/3)\,N_2 + H_2O(G)$	298	-226	-253.9	89
$(1/2)N_2H_4+(1/2)O_2 \longrightarrow (1/2)\,N_2 + H_2O(L)$	298	-312	-317.9	98.14
$C + O_2 \longrightarrow CO_2$	298	-394	-403.8	97.5
$\longrightarrow CO_2$	773	-396	-397.5	99.6
$\longrightarrow CO_2$	1273	-396	-397.2	99.6
$CO + (1/2)\,O_2 \longrightarrow CO_2$	298	-257	-282.6	90.9
$\longrightarrow CO_2$	773	-215	-283	75.9
$\longrightarrow CO_2$	1273	-172	-281	61.2
$(1/2)CH_3+(1/2)O_2 \longrightarrow (1/2)\,CO_2 + H_2O(L)$	298	-704	-726	96.9
$(1/2)CH_4+O_2 \longrightarrow (1/2)CO_2 + H_2O(L)$	298	-802	-803	99.8
$+H_2O(G)$	773	-800	-801	99.8

TABLE 2

THERMODYNAMIC EFFICIENCY OF

$H_2(g) + 1/2\,O_2(g) \longrightarrow H_2O(g)$

Reaction at Various Temperatures [2]

Tempera- (°K)	ΔG° (KJ/mole)	ΔH° (KJ/mole)	Efficiency ideal (%)	Carnot efficiency (%)
298	-229	-242	94.5	0
500	-218	-243	89.9	40
750	-205	-245	83.8	60
1000	-192	-247	77.8	70
2000	-134	-251	53.6	85

Carnot efficiency for an engine that operates between the given temperature, T_H, and a sink temperature of T_c = 298°K can be calculated from

$$\eta_{carnot} = 1 - \frac{T_c}{T_H} \qquad \ldots\ (11)$$

It can be seen from Figure 2 that the hydrogen-oxygen reaction would provide a fuel cell system potentially far superior in efficiency to any conventional heat engine system at temperatures approximately below

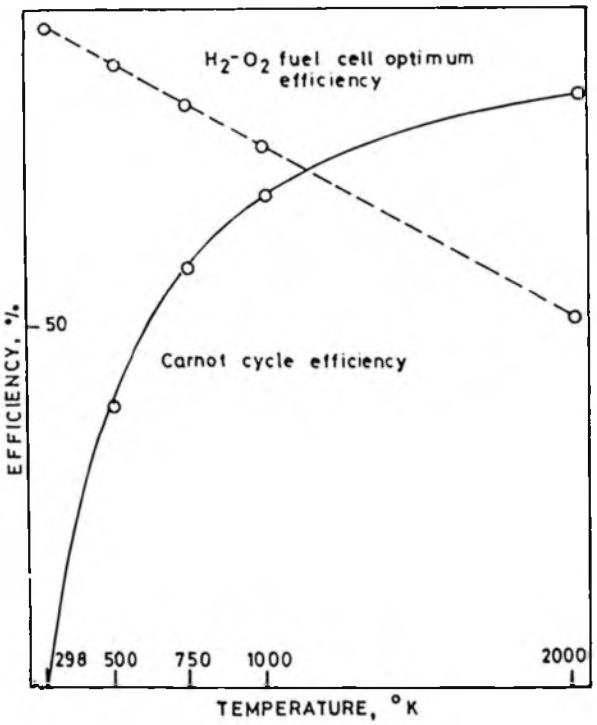


Fig. 2 Comparison of Carnot Cycle Efficiency and ideal efficiency of hydrogen - oxygen fuel cell.

about 1100°K. The discussion of thermodynamic efficiency defined by equation (10) is based on equilibrium conditions.

The performance of a fuel cell is represented by the current density versus voltage or polarization curve. When a net current is drawn to drive an external load, the cell voltage reduces to a value less than the thermodynamic equilibrium value, E, calculated from the Gibbs free energy change (equation 6). For example, for the hydrogen-oxygen cell at 250°C with the gases at atmospheric pressure, the ideal emf is 1.23 Volts. For the moderate currents at which fuel cells operate, the emf is 0.7 to 0.8 volts The voltage losses from ideal are referred to as polarization (Figure 3) and are generally attributed to one or more of the following:

(1) A hindrance to the reaction at the anode or cathode (called activation or chemical polarization);

(2) A concentration gradient in the electrolyte or in the reactants;

(3) Ohmic or resistance polarization.

The actual cell voltage can be written as:

$$V = E - \Delta V_{chem\,(c)} - \Delta V_{chem(a)} - \Delta V_{conc\,(c)} - \Delta V_{conc\,(a)} - \Delta V_{conc\,(g)} - \Delta V_{ohmic} \quad \ldots (12)$$

Where E is the ideal potential, as defined by Equation (6) and subscripts c and a denote cathode and anode respectively. The magnitude of each type of potential losses increases with current density as illustrated in Figure 3. Therefore, the cell potential and the efficiency of the cell decrease with increasing current density.

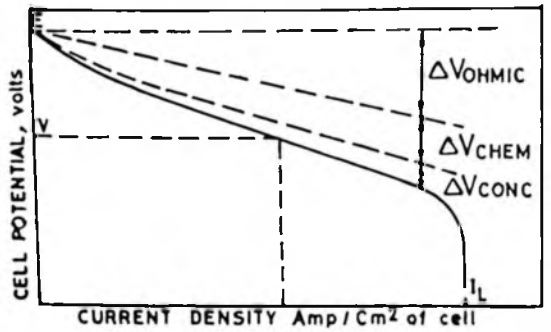


Fig. 3 Typical Cell Potential and Efficiency vs. Current relation of a fuel cell, showing regions of influence of various types of over-potential losses

Activation Polarization

Most chemical reactions involve an energy barrier that must be overcome by the reacting species. The activation energy results in activation polarization, which is primarily related to the slow step in the reaction sequence. The slow step could be adsorption of reactant onto the surface, electron transfer, desorption of product, or any other step in the reaction sequence. The magnitude of activation polarization, therefore, depends in part upon how ions are discharged at the electrodes and in part upon the rate at which they are discharged. The voltage drop due to activation polarization is related to the current density by an empirical equation, called the Tafel equation [3,4].

$$\Delta V_{chem(a)} = a + b \ln J \quad \ldots (13)$$

where J is the current density at the electrode; a and b are given

$$a = b \ln J \quad \ldots (14)$$

and

$$b = -\frac{RT}{\alpha nF} \quad \ldots (15)$$

The quantities α and J_O are kinetic parameters, the former being a constant that represents the fraction of ΔV_{chem} that aids a reaction in proceeding, the latter being the exchange current density, intimately related to the height of the activation energy barrier. Gas diffusion electrodes which maximize the three-phase interface of gas-electrode-electrolyte have been developed to reduce chemical polarization [5]. Also, numerous electro catalysts have been tried to achieve the same end chemically [6]. The activation polarization can alternatively be minimized by increasing operating pressure and temperature [1].

Concentration Polarization

Concentration polarization occurs when the electrode reaction is hindered by mass transport effects. The controlling process in the case of concentration polarization may be slow diffusion in the gas phase through the pores of electrodes, solution/de solution of reactants/products into/out of the electrolyte to a reaction site. At practical current densities, the latter predominates. A concentration gradient set up in the electrolyte produces a back emf which opposes the voltage that a fuel cell would deliver under

completely reversible conditions. When the electrode is completely governed by diffusion processes because of low reactant concentration in the feed gases or because of conversions approaching 100% (reactants depleted), the limiting current, J_L , is reached. This situation is characterized by a rapid drop in cell voltage, as shown in Figure 3. At the anode, where ions are being discharged from the electrode into the electrolyte and a concentration build up occurs, the concentration potential is given by [2].

$$\Delta V_{conc(a)} = \frac{RT}{nF} \ln \left[\frac{J_L + J}{J_L} \right] \quad \ldots(16)$$

For a consumptive electrode process such as at the cathode, where ions are being removed from the electrolyte, the concentration potential is given by

$$\Delta V_{conc(c)} = \frac{RT}{nF} \ln \frac{J_L}{J_L - J} \quad \ldots(17)$$

It should be noted that $\Delta V_{conc\,(a)}$ is usually small compared to $\Delta V_{conc(a)}$ as the logarithmic term in equation (16) can never exceed ln 2 as J approaches J_L For this reason, major efforts have been made to improve cathode or the oxygen electrode performance. The concentration polarization can be reduced most effectively by vigorous stirring of electrolyte and by increasing the operating temperature, both of which tend to accelerate ionic diffusion within the fuel cell. The last type of concentration polarization is due to changes in concentration of the reactant gases in the immediate vicinity of the reaction zone at the electrode. The gas pressure in the pores of the electrode must be sufficiently large to prevent the drowning of the electrode by the electrolyte, that is, it must be able to maintain three phase equilibrium within the pores of the electrode. The difference in pressure between the gas supply and the pressure found at the reaction zone causes a reduction in potential similar to the concentration gradient in the electrolyte

$$\Delta V_{conc\,(g)} = \frac{RT}{nF} \ln \left[\frac{P_r}{P_g} \right] \quad \ldots (18)$$

where P_r is gas pressure in the pores and P_g is bulk gas pressure. In general, this loss is small and can be neglected if pure reactant gases are used. However, for a gas mixture, gas concentration polarization may be significant. For example, if the oxidizer is air then P_g must be the partial pressure of the oxygen in the air while the electrolyte may be at the pressure of the air.

Resistance Polarization

When an anode and cathode are connected to an external load, the circuit is complete and current flows. Resistance to conduction of ions through the electrolyte, to electrons through the electrodes and current collectors, and by contact resistances between components causes ohmic or resistance polarization. Because both fuel cell electrodes and the electrolyte must obey Ohm's law, the voltage gradient required to force ionic and electronic conduction is given by:

$$\Delta V_{ohmic} = IR \quad \ldots (19)$$

Where R is the total cell resistance including electronic, ionic and contact resistances. The Ohmic losses can be reduced by using thin electrolytes and by efficient contacts between electrode and leads. Temperature and electrolyte composition also influence the cell potential. As temperature increases, resistance to electronic conduction in metals increases, however, resistance to ionic conduction in the electrolyte decreases. This results in a net decrease in cell resistance if the electrolyte concentration remains constant. The voltage efficiency is defined as the ratio of the actual cell voltage, V, underload conditions as calculated from equation (12) to the theoretical voltage at equilibrium:

$$\eta_v = \frac{V}{E} \quad \ldots (20)$$

The efficiency of an electrochemical cell is further reduced if all of the reactants are not reacted electrochemically to give current. The amount of current produced by an electrochemical reaction is obtained from Faraday's law [1].

$$I_F = nF \left[\frac{df}{dt} \right] \quad \ldots (21)$$

where (df/dt) is the molar flow rate of the electrochemical fuel to the fuel cell. The current or faradic efficiency is the ratio of the actual current to the current predicted by equation (21):

$$\eta_F = \frac{I}{nF\ (df/dt)} \qquad \ldots (22)$$

The quantity $(1 - \eta_F)$ is the fraction of the reactants that may react directly to release heat in the cell or may react to products other than those desired. The electrochemical efficiency of a fuel cell is the product of the efficiency terms that have been developed above.

$$\eta_E = \eta_T\ \eta_V\ \eta_F \qquad \ldots (23)$$

This equation is valid for a fuel cell operating on a gas containing inert and electrochemically active species, such as hydrogen. However, the presence of conventionally combustible species require an additional efficiency term.

A fuel cell power plant system consists of a fuel processor that produces H_2 rich fuel from raw and primary fuels, a power section that converts the chemical energy in the gaseous feeds to d.c power, and a power conditioner that convents d.c to a.c power. The heat dissipation or recovery function is satisfied by a Thermal Management subsystem. These subsystems are presented schematically in Figure 4.

The fuel cell subsystem is the heart of a fuel cell power plant and consists of :

(1) A plurality of single cells assembled in a stack;
(2) Manifold, ducting, blowers, heat exchangers and other accessories to distribute the fuel and oxidant gases at the desired temperature and pressure;
(3) Means for heat removal from the stack to maintain a steady operating temperature.

The fuel processor subsystem, as shown in Figure 4, may comprise of one or more of the components, depending on the fuel. Because the fuel converter may be sensitive to impurities such as sulphur and chloride compounds, these impurities must be removed. A variety of fuels as described in the subsequent section may be used. Conversion of the fuel to hydrogen rich gases may be accomplished by steam reforming, partial oxidation or cracking. In lower temperature fuel cells, for whom carbon monoxide is an anode poison, the concentration of carbon monoxide is minimized by the water shift reaction, or sometimes by methanation. Hydrogen rich gases are fed to the fuel cell. Because water is required in the fuel conversion processes and is produced in the fuel cell reaction, the required water may be recovered from the exhaust gases. Depleted gases from the fuel cell subsystem may also be recycled to the fuel converter or burned to provide for the endothermic heat or reaction.

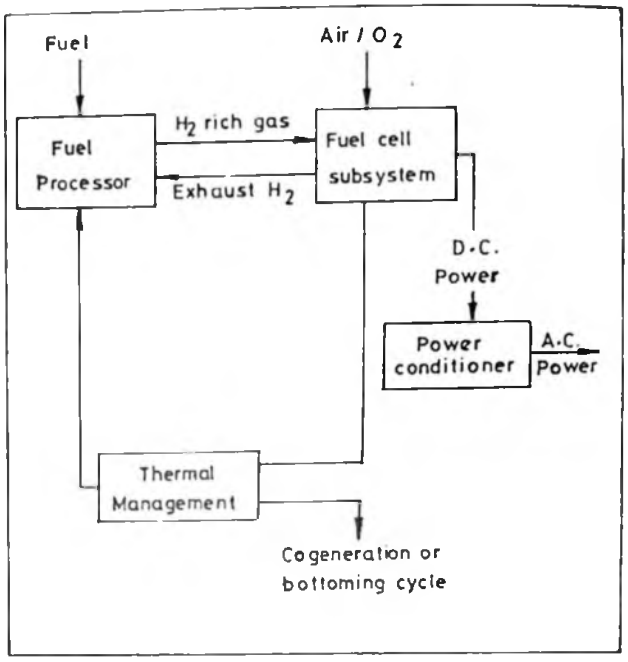


Fig. 4 Schematic of Integrated Fuel cell System.

The product gases from most practical fuel processor will contain varying amounts of inerts and combustibles like H_2, CO and CH_4, which can be burned in a conventional combustion device, converting all chemical energy into heat energy usable in a heat engine. The fuel cell converts only H_2 directly and, at high temperatures, CO and CH_4 indirectly to electricity. The heating value efficiency is defined as the ratio of the lower heating value of the electrochemically active fuel, ΔH_r, to the lower heating value of the fuel gas containing all combustible species.

$$\eta_H = \frac{\Delta H_r}{\Delta H_c} \qquad \ldots (24)$$

The fuel cell subsystem efficiency describes the conversion of chemical energy in the feed gas to d.c power and is obtained by combining equations (23) and (24)

$$\eta_{FC} = \eta_E \eta_H \quad \ldots (25)$$

The overall efficiency of a system without waste heat utilization is

$$\eta_S = \eta_{FP} \eta_{FC} \eta_{PC} \quad \ldots (26)$$

Where η_{FP} is the fuel processor efficiency, given by the ratio of the low heating value of gaseous fuel from fuel processor to fuel cell to the high heating value of raw fuel into fuel processor.

Fuel Cell Heat Balance

The heat balance of a fuel cell can be determined from the same concepts used to develop the efficiency equation. Figure 5 and Equation 27 summarize the heat balance of fuel cell.

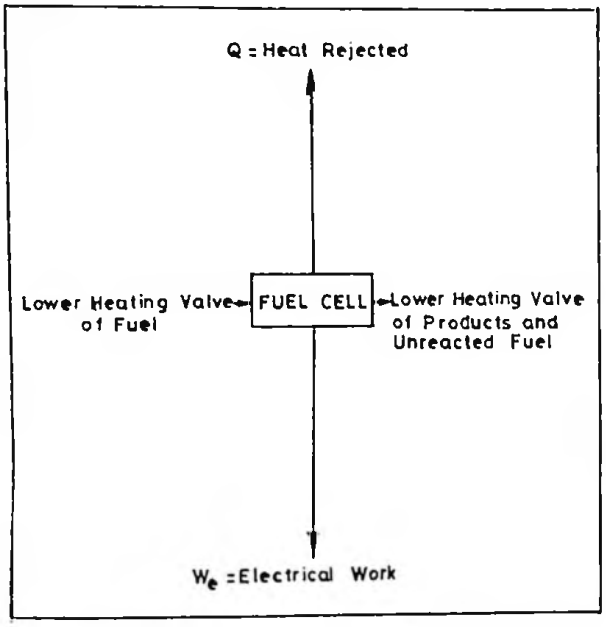


Fig. 5 Fuel Cell heat balance.

The lower heating values of the inlet and exit streams are easily calculated if the composition are known:

$$LHV_{in} = W_{el} + Q + LHV_{out} \quad \ldots (27)$$

The electrical work obtained is related to the electrochemical efficiency of a fuel cell by equation 28:

$$W_{el} = \eta_E \Delta H \quad \ldots (28)$$

The heat released, Q within a fuel cell comes from three distinct effects taking place in the cell:

(1) The electrochemical reaction that gives rise to the reversible heat transfer;

(2) The fuel reacting chemically with the oxidizer rather than electrochemically to generate an irreversible heat transfer;

(3) The voltage drop manifesting itself as I^2R or $I \Delta V$ heat in the cell.

The reversible heat transfer is obtained only from those moles of fuel that react electrochemically to produce an electric current, namely η_F (df/dt). Thus, the reversible heat transfer rate is:

$$\begin{aligned} Q_1 &= \eta_F \ (df/dt) \quad (T \Delta S) \\ &= \eta_F \ (df/dt) \quad (\Delta H - \Delta G) \\ &= -\frac{I}{nF} - (\Delta H - \Delta G) \end{aligned} \quad \ldots (29)$$

Where, η_F (df/dt) has been eliminated in favour of the current by use of equation (22). Since $T \Delta S$ term for most reactions of interest is negative, the reversible heat transfer in most cases will be away form the cell.

The part $(1 - \eta_F)$ of the fuel supplied that reacts chemically, but not electrochemically with the oxidant also turned into heat, this amounts to:

$$\begin{aligned} Q_2 &= (1-\eta_f) \ (df/dt) \ (\Delta H) \\ &= \frac{I}{nF} \left[\frac{I-\eta_F}{\eta_F} \right] (\Delta H) \end{aligned} \quad \ldots (30)$$

Since ΔH is also negative for most fuel cell reactions of interest, this results in heat transfer away from the cell. However this term is generally small, since η_F is usually close to one.

Finally, the heat transfer also takes place because of the irreversible voltage drop that occur in the fuel cell. As discussed earlier, the cell operating voltage is below the theoretical open circuit voltage due to activation and concentration polarization at each electrode as well as I^2R type voltage drop occuring at each electrode and in the electrolyte. Considering the fuel that reacts electrochemically to produce an electrical current

$$Q_3 = \eta_F (df/dt) n_e (V - E) = I (V - E) \quad \ldots (31)$$

The total heat transfer is then

$$Q = Q_1 + Q_2 + Q_3 \quad \ldots (32)$$

FUEL CELL TYPES

In the course of more than 140 years of development, various types of fuel cells have been developed, based on a number of parameters which are listed in Table 3. Of all the parameters mentioned, temperature and electrolyte have been the governing factors in the development of fuel cells. Ever since the fuel cells were developed, there has always been an attempt to increase the operating temperature of the system, as it overcomes the various kinds of overpotentials (mentioned in the previous section), which eventually decrease the efficiency of the system. Increase in temperature enhances the rate of electrochemical reaction, by decreasing the activation overpotential as is evident from equations (13,14,15) of section 2. In addition, high temperature also accelerates mass transfer process in the system, i.e., increases the diffusion coefficient of the reacting gases through the electrode as shown by the following equation:

$$D \propto T^{3/2} \quad \ldots (33)$$

where D is the diffusion coefficient of the reacting gases and T is the temperature of the operating system. Increased temperature also hastens the diffusion of the reacting ions through the electrolyte to the reaction site and further overcomes the concentration overpotential in the system. Besides, cell resistance decreases considerably with the rise in temperature.

The other parameter, electrolyte, is of equal importance but the choice of electrolyte had always been temperature dependent. At the outset, among the aqueous electrolytes for low temperature systems, various alkaline electrolytes had been tried. The main disincentive to work with alkaline electrolyte has been the formation of less conducting bicarbonate ion when the hydroxyl ion from the alkali reacts with the main reaction product, carbondioxide as follows:

$$OH^- + CO_2 \longrightarrow HCO_3^- \quad \ldots (34)$$

The use of acid electrolytes avoids these complications as it can tolerate carbonaceous fuel as well as CO_2 in the oxidant gas. Among the acids commonly available, hydrochloric acid is rejected [7] as it easily loses hydrogen chloride gas when other gases are bubbled through it or come in contact with it in the electrode pores. Nitric acid has too great an oxidising power and consequently is very damaging to the cell structure and the electrode. Other acids are either too little dissociated or too expensive. Sulphuric acid may be used as an electrolyte because of its higher conductivity. However, it is highly corrosive, viscous when concentrated and has a high heat of dilution. It has considerable oxidising power and may yield on reduction hydrogen sulphide, a serious catalyst poison. Phosphoric acid, despite its comparatively low dissociation constant, is currently most common. Solid polymer acidic electrolyte has also been tried, which uses ion exchange membrane as the electrolyte [8]. The electrolyte being a solid, cannot change, move about or vapourize from the system, thus has an advantage over other electrolytes. Though the electrolyte loss is controlled, it has application in a limited temperature range.

TABLE 3

FUEL CELL PARAMETERS [8]

Fuel		Oxidant	Temperature (°C)	Electrolyte
Direct	Indirect			
H_2 Hydrazine	Hydride Ammonia	Oxygen Air	Low(260) High (600-700)	Aq. Acid H_2SO_4
Ammonia	H-C	H_2O_2	V.High (>750)	Phosphoric Acid
H-C	Methanol			Aqueous Alkaline: NaOH, KOH
MeOH Coal Gas	EtOH Coal Gas			
Coal	Coal			Solid Polymer
				Molten Carbonate
				Solid Oxide

Here electrolyte being solid in nature must be hydrated to perform. Therefore, cell must be operated under such conditions where the by-product water does not vaporize in the reaction air stream faster than it is produced during the electrochemical reaction. This constrains cell operation to under 60° C at ambient pressure and about 120°C at elevated pressure. Moreover, solid polymer electrolyte freezes at about 0°C. In general, aqueous electrolytes cannot be used at higher temperatures as there is always a danger of electrolyte loss by evaporation; at the same time these electrolytes may corrode the cell structure at high temperatures; therefore, there was a need to search for alternative electrolytes, which can provide conducting species other than H^+ or OH^- ions, as aqueous electrolytes were found unsuitable. The best choice was contemplated to be oxide ions (O^{2-}) which can be produced at the cathode as follows [7]:

$$O_2 + 4e^- \longrightarrow 2O^{2-} \quad \ldots (35)$$

Ions thus produced move to the fuel electrode to oxidise the hydrogen and hence conduct electricity.

$$H_2 + O^{2-} \longrightarrow H_2O + 2e^- \quad \ldots (36)$$

Molten ionic oxide was thought to provide the best electrolyte to encourage this process. However, simple ionic oxides have melting points greater than 1000°C and therefore, attention was focussed on salts, melting at lower temperatures. These salts were generally those with O_2 containing anions, e.g., nitrate, sulphate and carbonate. Since CO_2 is the major product in a fuel cell, it may react with any molten salt containing ions other than carbonate, displacing the appropriate acid and anhydride (SO_3) which may then attack the electrode material or be otherwise troublesome.

$$CO_2 + SO_4^{2-} \longrightarrow CO_3^{2-} + SO_3 \quad \ldots (37)$$

Hence, it is most satisfactory to consider a molten carbonate or mixture of carbonates as electrolyte. A mixture of salts may have a considerable advantage since it will have a lower melting point than either component on its own. So far the best system found is a mixture of lithium potassium carbonate and inert powder of lithium aluminate which is stable upto 700°C. At still higher temperatures i.e. 1000°C, solid oxide electrolyte was thought of as a suitable choice. At these temperatures, oxide ion thus produced at the cathode (as discussed earlier) can migrate through the vacant sites in the lattice of solid electrolyte to the anode, to oxidise the fuel (hydrogen) and thus conduct electricity by ionic mechanism. In this regard yttria stabilized zirconia $(Y_2O_3)_{0.15}$ $(ZrO_2)_{0.85}$ has been found to be very promising [9]. As there are no liquids involved, the problem like corrosion of the cell structure faced in earlier cases is absent.

Fuel cells can be classified by coupling the electrolyte system with temperature based upon the electrolyte's thermal capability as follows.

Low temperature (< 260°C)	Alkaline fuel cell (65°C - 160°C)
	Solid polymer fuel cell (< 120°C)
	Phosphoric acid fuel cell (175°C - 200°C)
High temperature: (600°C - 700°C)	Molten carbonate fuel cell
Very high temperature: (> 750°C)	Solid oxide fuel cell

The cells that have been most extensively developed are those that use hydrogen as their fuel and oxygen or oxygen in air as the oxidant. Figure 6 plots cell voltage, a measure of efficiency, against cell operating temperature for all fuel cell types that are currently being developed. A perfectly efficient fuel cell is shown for comparison. Even though phosphoric acid cell has the lowest efficiency of the fuel cell types shown, it is technologically the most advanced. It is considered as the first generation fuel cell and has the best combination of proven cell life, tolerance to impurities in the fuel and oxidant and low cost. Molten carbonate fuel cell is considered as the second generation fuel cell, while solid oxide fuel cells are being developed for the long term. Alkaline and solid polymer electrolyte fuel cells have only limited application where carbonaceous fuels or air are used as reactants. The important applications (in space and under sea), require pure hydrogen and oxygen.

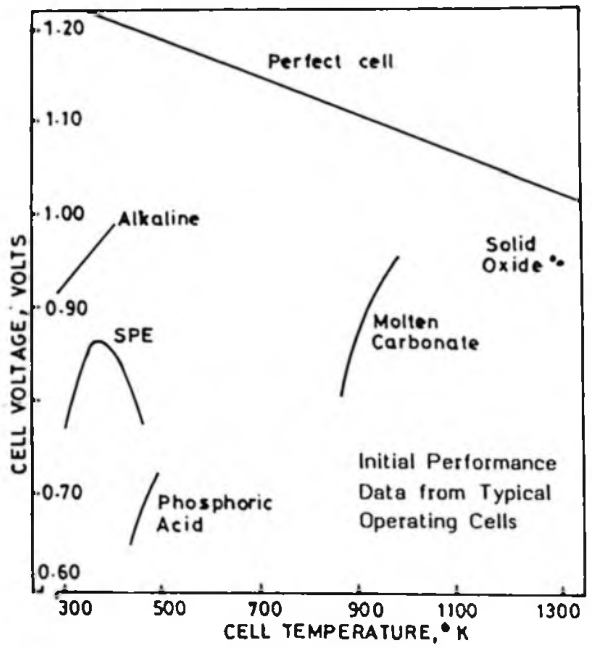


Fig. 6 Cell voltage vs. operating temperature for different fuel cell types.

STATUS OF FUEL CELL TECHNOLOGY

The development of fuel cells by research organizations and industrial firms has been underway mainly in the U.S.A., Japan and Europe. Efforts are practically devoted to phosphoric acid and high temperature fuel cell in the U.S. and Japan, but virtually nothing is done on alkaline and solid polymer electrolyte fuel cells. On the other hand, most European efforts so far have been focussed on Alkaline fuel cells.

Efforts in United States of America

Various organizations in the U.S. have been persuing the development of this technology, mainly on PAFC, since the mid sixties. As the design and engineering work has completed by early 1980's, it was decided by Gas Research Institute (GRI), Department of Energy (DOE) & United Technologies Corporation (UTC) to have field test and demonstration of this technology. It was carried along two lines:

(a) Power plants for on-site applications
(b) Utility power plants.

Since 1983, fifty of 40 KW PAFC on-site units are being installed for field tests at various places like restaurants, apartment complexes, etc., as a part of a programme which is expected to conclude by the end of 1986. The most successful of the field test according to GRI is the one in Portland, Oregon. It has logged over 3300 hours of successful operation [10]. In an effort parallel to the on-site field test, DOE, Electric Power Research Institute (EPRI) & UTC initiated in 1976 a 4.5 MW fuel cell power plant for demonstration [11]. It is built in Central Manhattan on a site provided by the Consolidated Edison Company of New York. So far it has not been successful and is running intermittently. Recognising the special value of the fuel cell in electric utility systems, UTC and Toshiba Corporation have formed an organization known as International Fuel Cell Corporation, located in South Windsor, Connecticut. The company is planning to install an 11 MW PAFC plant, which will incorporate certain improvements based on the experience of 4.5 MW demonstrator programme. The power plant is designed to operate on both natural gas and liquid hydrocarbon fuel. More recently in 1982, Westinghouse and Energy Research Corporation (ERC) have decided to install a 7.5 MW PAFC power plant in Southern California by mid 1987 [12]. It will use air cooled DIGAS system instead of water cooling. At present, the activities related with the transition from demonstration to commercialization are being coordinated by the Fuel Cell User's Group formed in 1981 In addition to first generation fuel cell, studies and development of a technology base for high temperature, second generation fuel cell power plant have been initiated by EPRI, UTC, DOE and GRI. The progress in fuel cell stack technology in the U.S. has been steady, albeit slow. There has been an order of magnitude increase in demonstrated stack durability over the past twenty or so years (Table 4).

TABLE 4

TWENTY YEAR PROGRESS IN FUEL CELL TECHNOLOGY (ORDER-OF-MAGNITUDE NUMBERS) [33]

	Early 1960's		Present Time	
	Largest Stack (KW)	Stack Life (hrs.)	Lagest Stack (KW)	Stack Life (hrs.)
Solid Polymer	10^{-1}	10^{2}	10	10^{4}
Alkaline	10	10^{3}	10	10^{4}
Phosphoric Acid	1	10^{3}	10^{3}	10^{4}
Carbonate	10^{-1}	10^{2}	1	10^{3}
Solid Oxide	10^{-2}	10^{2}	10^{-1}	10^{3}

Work in Japan

Research and Development on Fuel cells in Japan is supported as a national project, mainly by the government agencies like, Ministry of International Trade and Industry (MITI), and Agency of Industrial Science and Technology (AIST). Besides, private companies are also energetic. This programme which is included in the national 'Moonlight Project' was initiated in 1981 and has progressed successfully [13]. As in the U.S., more emphasis was laid on PAFC, due to its inherent good qualities as discussed earlier. The present major fuel cell power plant project has been completed by the Tokyo Electric Power Company in the Goi thermal power station campus at Chiba-Ren [14]. The Goi unit ordered from UTC ran for the first time in April, 1983 and after some maintenance work in 1984, it has been operating satisfactorily to produce 4.8 MW as designed. Upto March 1985, plant had run for 1900 hours and had produced 4200 MWh. It uses natural gas to generate electricity with an efficiency of 80% with waste heat recovery and of 40% without. The Tokyo Gas Company and the Osaka Gas Company participated in the Target project in 1972 and both companies started field tests of on-site integrated energy systems of PAFC PC-18 in March 1982. In addition to these 40 KW units, two 50 KW air cooled systems are supplying electricity and hot water to a factory. Two different types of 1 MW power plants are under construction for both dispersed and centralized electric power generation, which are ready for test operation by the end of 1985. As the demonstration work is over, it is expected that the PAFC will be commercialized in the electric power industries market around 1991. At the same time, efforts are now directed on the next stage of R&D on PAFC, to further improve the existing technology. Unlike PAFC, molten carbonate fuel cell technology has not matured as yet in Japan and is still in the process of development. Fundamental studies on materials have been made by manufacturers, national laboratories, universities, and is still an ongoing activity. Two models of 1 KW MCFC stack were tested and the design of a 10 KW model were to be completed by the end of 1985. At the same time feasibility studies on internal reforming cell technology are being carried out which is expected to be a well established technology within a few years. Other technologies like solid oxide, and alkaline fuel cells are still in the stage of infancy. R & D on SOFC is at the stage of fundamental research; a 500W stack of SOFC will be assembled this year. Alkaline fuel cell of 1 KW capacity was assembled and tested for over 2000 hours. The main thrust of R & D activity here, was on decreasing the loading of platinum electro-catalyst and searching cheap and suitable alternative for it.

Work in Europe

Fuel cell development in Europe have been much more modest in comparison and mainly concentrated on the Alkaline Fuel Cell, since Europeans have so far been interested in the use of fuel cells only for transport.

Siemens in West Germany have been pursuing the development of the H_2-O_2 alkaline fuel cell since the midsixties. A report [15] mentioned the testing of three 7.5 KW fuel cells in a laboratory simulation of a hospital standby power system which includes lead acid batteries for startup.

Elenco, a Belgium/Dutch consortium founded in 1976 by three stockholders (two from Belgium, one from Netherlands) has adopted a pragmatic approach in the field of developing and implementing fuel cells in the commercial vehicle market. It has chosen the hybrid power source approach so as to be able to use the fuel cell mostly at constant power level. Elenco is also involved in the scaling up of its fuel cells and the largest one operated so far is the 12 KW fuel cell of the van. At present, a 40 KW cell is under construction for the Belgian Geological Service which plans to use the system to provide power for its drilling and measurement equipments at all possible underground sites in Belgium [16].

In Italy, work on acid fuel cell for transport has been carried out at the Fiat Research Centre. Test results with a Fiat 242 electric van have demonstrated that an integrated system of metal hydride storage and fuel cells characterised by an energy density of 140 Wh/kg, can perform successfully for 3h at a top speed of 70 Km/h. Recently, the research attention is also directed towards preliminary material studies for MCFC electrodes, processes for SOFC and high protonic diffusivity.

Though no major work has been done in UK, two distinct fuel cell applications have been identified: transport, which requires fuel cells that work at temperatures below 100° C and base-load power generation utilizing coal gas. The Central Electricity Generating Board studies have indicated that in the post 2000 era, an intermediate temperature (600°C-800°C) fuel cell operating in a coal-fuelled integra-

ted system could provied an attractive option for power generation. Research activities to meet the requirements in the time frame available are in the stage of planning [17]. In Denmark, the Energy Research Laboratory (ERL) and the Odense University are engaged in a collaborative project on the development of advanced solid state fuel cells [18].

Indian Scene

Fuel cell research in India is still in an incipient stage and is so far directed towards basic research. Little efforts have been made in applied research. There are not many groups in India currently engaged in this technology though the first work was initiated in 1974 by the researchers at Central Fuel Research Institute (CFRI), Dhanbad. Mukherjee and coworkers [19] investigated the techniques for the fabrication of catalyst impregnated carbon electrodes having proper pore size distribution for low temperature H_2-O_2 fuel cell. Rao and coworkers [20] of the Indian Institute of Technology, Kharagpur studied the electrochemical oxidation of formamide in acid medium at platinized platinum, platinum-palladium, platinum-tungsten, platinum-nickel and platinum-gold electrodes using galvanostatic method. They found the catalytic activity of platinum-gold alloy electrode better than others with maximum catalytic activity obtained from 35% gold alloy. Subrahmanyan and coworkers [21] of Indian Space Research Organization (ISRO), Satellite Centre, Bangalore, have reviewed this technology for space application. They have examined its applicability in space upto the turn of the century and identified the research areas. Shukla and coworkers [22] of the Indian Institute of Science, Bangalore have worked mainly on low cost electrodes. They fabricated porous air electrode based on coconut shell charcoal which could obtain 0.5-2.0 Amp/cm^2 for 200h in alkaline electrolyte. Its current density was further improved to 2-5.2 Amp/cm^2, by loading the electrode with 7 wt% of Platinum and to 0.001 Amp/cm^2 with non noble catalyst like Perovskite oxide. In addition to alkaline electrolyte, acidic electrolytes were also tried. 2.5M H_2SO_4 and 7M H_3PO_4 could sustain currents upto 600 mAmp/cm^2 at a polarization upto 0.4V from their rest potential. Venkatesan and coworkers [23] of the Central Electrochemical Research Institute, Karaikudi have developed 1 kW low temperature alkaline H_2-O_2 fuel cell, containing skeletal nickel and silver catalysts for the hydrogen and oxygen electrodes respectively. They have fabricated and tested single cells consisting of electrodes of three layer DSK type and having area of 15 cm^2 (SC-I) and 30 cm^2 (SC-II). The SC-I showed a cell voltage of 0.70 V at current density of 0.035 Amp/cm^2 at 60°C while the SC-II showed a cell voltage of 0.75 V at current density of 0.015 Amp/cm^2 at 50°C. They have also assembled 30 fuel cell modules consisting of two cells, three cells, four cells or six cells. A number of two, three and four cell modules tested at room temperature showed a satisfactory performance for periods ranging from 2000 hours to 7500 hours at current densities in the range of 0.015 to 0.30 Amp/cm^2. Notable contributions have been made by Govil and coworkers [24-28] of the Tata Institute of Fundamental Research (TIFR), Bombay in the area of biochemical fuel cell. The biochemical fuel cell is a device which converts the chemical energy of a hydrogen rich fuel such as alcohol, glucose, hydrocarbons or hydrogen itself into electrical energy through enzyme catalyzed oxidation reduction reactions. Banerjee, [29] at the University of Delhi has initiated a research programme to investigate the suitability of various low temperature H_2-O_2 fuel cell for low cost power supply. Vasudevan and coworkers [30] at the Indian Institute of Technology, New Delhi have reviewed various classes of electrocatalysts for H_2-O_2 and hydrocarbon fuel cells. They are exploring future directions in synthesizing more efficient electrocatalysts for the successful operation of the fuel cells. Tata Energy Research Institute, New Delhi has proposed to develop an experimental single fuel cell for electrode kinetic investigations of fuel cell reactions. It also plans to carry out a site specific techno-economic analysis through demonstration project to assess feasibility of fuel cell technology. It is felt that only through such detailed evaluation will specific areas be revealed wherein major modifications, improvements and economies can be brought about to make the use of fuel cell technology a viable alternative in India.

FUEL CELL APPLICATIONS AND R & D NEEDS IN INDIA

Most people had never heard of a fuel cell, until 1960s when the first manned spaceflight programme took off. This technology attracted attention of technologists for terrestial applications only after 1973 oil embargo, when alternative sources of energy were searched for. High efficiency was one of its unique characteristics which aroused interest in scientists and engineers. Furthermore, the efficiency is essentially independent of size.

Small power plants operate nearly as efficiently as larger ones even at part load. Apart from this, fuel cells have shown to possess a number of important advantages over any other type of generator now available. Modular nature is one of the most significant of them. They can be stacked and put together in virtually any configuration, almost any voltage and power can be generated without the loss of their desirable characteristics. Moreover, since it essentially avoids combustion process, fuel cells do not pose a threat of environmental pollution, unlike the conventional power plants. A comparison of emissions from conventional power plants with those from an experimental fuel cell working on natural gas shows the measured emissions of NO_x, SO_x, smoke and particulates from the fuel cell 1/10th to 1/50,000th of those of conventional electric generators [31]. As discussed previously, fuel cells can use any two chemicals that can undergo an oxidation reduction reaction. However, the most practical fuel for a fuel cell remains hydrogen. Fuel cells can therefore, use a variety of renewable fuels like natural gas, methane, methanol, medium Btu gas from coal gasification, propane, naptha, as all of them yield hydrogen on requisite reforming. Hence the technology will not become redundant if one type of fuel is not available. Heat generated during the electrochemical reaction in the fuel cell can be recovered without reduction in the system's efficiency. In addition, it shows almost instant response to changes in power demand from minimum load level to maximum level in approximately 30 milliseconds. These qualities are of importance, especially in Indian context, where present preoccupation is with saving of energy resources and the concern for protection of the environment.

One of the significant advantages that fuel cells are capable of providing in terms of power generation capability is its compatibility with other renewable technologies such as solar cells, wind power generators, hydroelectric generators, and other similar systems. All solar cells generate direct current, as do many wind systems. Both are intermittent generators and need an additional source of power to provide electricity when the motive force (wind or sun) is not available. Both can be directly compatible with fuel cells by choice of proper design and by matching of voltages. Many wind turbines have complicated mechanisms and/or electronics to allow them to match AC system frequencies and to synchronize voltage and power curves. A DC system does not need to be synchronized and only needs a rectifier to prevent backflow of current. As a result, less expensive wind turbine systems could be utilized. Batteries for storage are also, of course, compatible with fuel cells. Fuel cells could be used to charge batteries for surge capability or to provide reserve when powerplant capacity is exceeded. This would allow the purchase of smaller fuel cell capacity. Much can be said for the use of direct current power systems when considering the number of new renewable energy systems now becoming available.

The demand for power in the country has exceeded its availability causing widespread shortages in different regions. The 12th Electric Power Survey of India Report [32] indicates that the installed capacity for the country as on 31st March, 1985 was 42771 MW with the peak availability estimated around 26000 MW. Compared to this, the peak load was of the order of 31000 MW and in terms of energy, a deficit of 7769 MKWh, the deficit is likely to go upto 12400 MKWh in 1990. Reasons for this deficit are manifold; firstly, only 56% of the installed capacity of the power plants is utilised due to shortage in coal supply, age of the plant and poor plant management. Closer look at the thermal power plant performance in the country reveals that we generate roughly 75% of our electricity at just over 25% efficiency, which is a gross wastage of energy reserves in the country. It may be conserved, if highly efficiency technologies like fuel cells are put to use. Another reason for poor performance is that old thermal power plants are being operated inspite of their life being exhausted; replacement and construction of new plants are very time and capital intensive. This problem can be avoided by fuel cell power plants. Being modular, they can be factory produced within 18 months to 2 years time, compared to 5-7 years of gestation time needed for conventional power plants. Moreover, during plant operation, the modular concept would permit rapid replacement of defective parts. Furthermore, capacity expansion of an existing installation can be facilitated by adding modules without affecting the system's efficiency or initial cost. A recent report has revealed that better quality coal reserves have already been exploited, the leftover is poor quality coal which often leads to plant breakdown and further aggravates the energy problem. It is in this context, a major engineering change will have to be envisaged if the fuel is to be changed. On the other hand, fuel cells can easily adapt to different

types of fuels, and the technology will not become redundant. Besides the energy losses, thermal power stations are the largest source of emissions of a number of pollutants, while the environmental compatibility allows fuel cell power plants to be located in population centres and solve environmental as well as difficult transmission problems. Large generating stations operate most efficiently at a rated power output, but the demand for power is variable. Their efficiencies drop if not operated at the rated output, i.e. during non peak hours. A possible solution for this would be to operate the system at the full capacity, excess electricity could be stored through electrolysis as hydrogen and can be retrieved back to meet the peak demand by fuel cells. The transmission and distribution (T&D) network in the country is in a highly disordered state, mainly on account of lack of proper planning. Growth of T & D system has not matched with the expansion of power generation capacity. Increasing the capacity of lines to remote areas, away from the central grid, implies increased T & D cost. The investment costs for new T&D lines are between Rs. 20,000 - Rs. 30,000 per KW. At the same time, the existing T&D network suffers from the losses of the order of 20 - 25% of the energy delivered at busbar, while these losses are just 8-10% in developed countries. The factors contributing to these losses are (i) extension of the distribution network covering more and more areas of the state; (ii) low load deficit and long lines; (iii) introduction of flat rate tariff for agricultural consumers in many of the states; (iv) unauthorised and undetected use of energy. Hence, in a nutshell, less than 20% of the total supply caters to the 75% of the Indian population, residing in remote areas. Therefore, grid electricity must be viewed as a limited source for rural electrification. Here a decentralised power generation, therefore seems to be an appropriate choice. Looking back, fuel cells seems to be a suitable candidate. Being modular, it will combat the inherent ills of the central grid system. The need for electricity in Indian villages is so immediate, as the limited supply has hampered the agricultural productivity and the development of the country as a whole. Dispersed fuel cell power plant (10-200 KW), which does not require long gestation period, would definitely bring relief to the existing crisis. Significant and continued increase in the price of oil and also uncertainty in its availability further puts pressure on the country's economy. Besides costs, increasing environmental pollution has always led engineers to turn their mind for alternative system. Several types of available secondary batteries were envisaged to be suitable option. Infact, in our country, Bharat Heavy Electricals Limited (BHEL) has developed battery powered vehicles of 1 tonne payload using separately excited DC motor. These vehicles have been deployed during important events like Asiad-82, NAM-83, and also by members of Parliment. Though being competent, it cannot be adopted for long term continuous operation. Fuel cells seem to be the likely choice to solve the existing problems, being modular, pollution free and capability to adapt to various fuels. Efforts have already been started in Europe by Elenco Company. They have designed the hybrid power source viz. a combination of fuel cell and battery modules. Although fuel cells are highly efficient, they cannot provide excess power during acceleration, being low specific power device. In the hybrid system, a set of batteries overcomes the above limitation. This system in terms of weight, volume and cost meets the operational requirement very well. However, for the complete market diffusion, fuel cell technology has yet to undergo a significant maturation.

It is evident from the previous discussions that fuel cell research needs expertise from various disciplines. Hence, keeping that in view, a programme coordinated at national level may bring successful adoption of this technology in India. As reviewed earlier, this technology is in fairly advanced stage in developed countries like United States of America and Japan; it will be in our interest if we focus our efforts more on its plausible aplications in the country Our requirements are very different and the major need is for remote rural areas, as over the years,the cost of supplying electricity from the central grid to rural areas in India has gone up rapidly to Rs. 5/KWh. The prospect for the future is even more heavily loaded against centralized power supply, because a large bulk of villages yet to be electrified are further and further away from electricity. At this cost, decentralized systems like fuel cells could become serious competitors, as the major emphasis is now being laid on utilization of renewable forms of energy such as biomass. India produces annually 200 million ton of agricultural residue such as wheat straw, rice husk, nut shells etc. This biomass is mainly organic in nature, a substantial protion of which is cellulose, which can be converted into gaseous (methane) or liquid (methanol, ethanol) fuels by biological processes like anaerobic digestion and fermentation. This enzymatic break-

down of biomass at low pressure and low temperatures is accomplished by using specific microorganisms. In the context of a vast network of biogas plants in India, dovetailing of the fuel cell technology into the simple biogas system could mean a self sustained decentralized energy system. The alluring proposition of coupling of biofuels with fuel cells should be examined in India through actual engineering, installation and evaluation of such a system. It is only through such studies, it will not only be possible to determine the limits of economic viability with existing technology but will also be possible to identify areas for further research and development, necessary for the widespread diffusion of fuel cell technology in India. Besides the important considerations that presently exist for achieving commercial accepetance of fuel cells are centered on capital cost and lifetime. Thus, efforts should also be directed on some basic research which will focus on the development of alternative low cost catalysts, construction materials, reduction in expensive noble metal catalyst loading and improvement in electrode design. A test facility should be developed to study various electrocatalysts and electrode design, for their performance evaluation and kinetic measurements. Fundamental research should also be done to develop second generation fuel cells such as Molten Carbonate and Solid Oxide fuel cells.

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