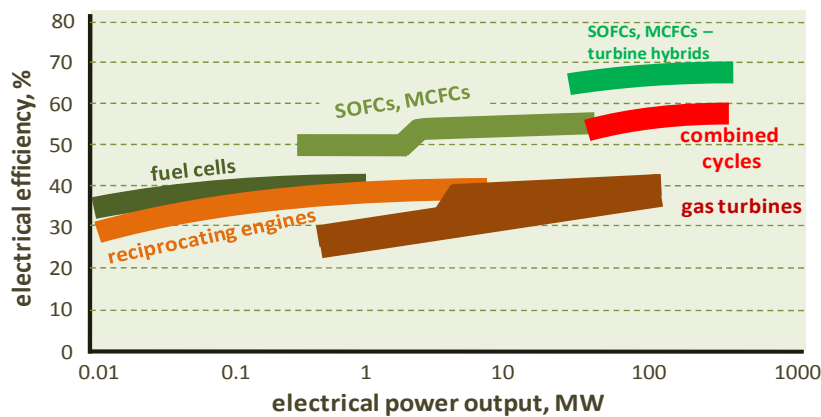


## 1. Introduction

Fuel cells are regarded as the technology of choice to maximize the potential benefits of hydrogen in terms of efficiency. Today's fuel cell plants exhibit efficiencies in the range of 40 to 55% LHV, almost independently of their size, while hybrid fuel cell – gas turbine cycles overcome 70% LHV (Figure 1). They are electrochemical devices that convert the chemical energy of a fuel directly to electricity, bypassing the thermodynamic limitations of conventional thermal engines. Their physical structure consists of an (solid or solidified) electrolyte in contact with two porous electrodes on either side. All types of fuel cells combine hydrogen and oxygen to produce dc electricity, water and heat. On the other hand, their sensitivity and endurance over time and above all their high initial costs are the main obstacles for broad commercialization [1-3].



**Figure 1** Comparative efficiency (% LHV) of power generation systems [1,2]

Based on the type of the electrolyte the most common classification of fuel cells includes:

1. proton exchanging membranes (PEM) fuel cells (or polymer electrolyte fuel cells – PEFCs), with proton conducting polymeric membranes, transports hydrogen (fuel) cations, generated at the anode, to an ambient air exposed cathode, where they are electro-oxidized to water at low temperatures
2. solid oxide fuel cells (SOFC), which use oxygen conducting ceramic membranes to electro-combust  $H_2$ , at the anode, by  $O^{2-}$  anions provided by the cathodic reduction of ambient oxygen at high temperatures
3. molten carbonate fuel cells (MCFC), with alkali carbonate (in  $LiAlO_2$  matrixes) electrolyte, conduct  $CO_3^{2-}$  anions, generated at an  $O_2/CO_2$  exposed cathode to electrooxidize  $H_2$  (fuel) at the anode and at high temperatures
4. alkaline fuel cells (AFC), with concentrated KOH (in asbestos matrixes) electrolyte, conduct  $OH^-$  anions, generated at an  $O_2/H_2O$  exposed cathode to electrooxidize  $H_2$  (fuel) at the anode and at moderate temperatures, and
5. phosphoric acid fuel cells (PAFC) with concentrated  $H_3PO_4$  (in silicon carbide matrixes) electrolyte, which transports  $H^+$  cations, generated at the anode, to an ambient air exposed cathode, where they are electro-oxidized to water at moderate temperatures

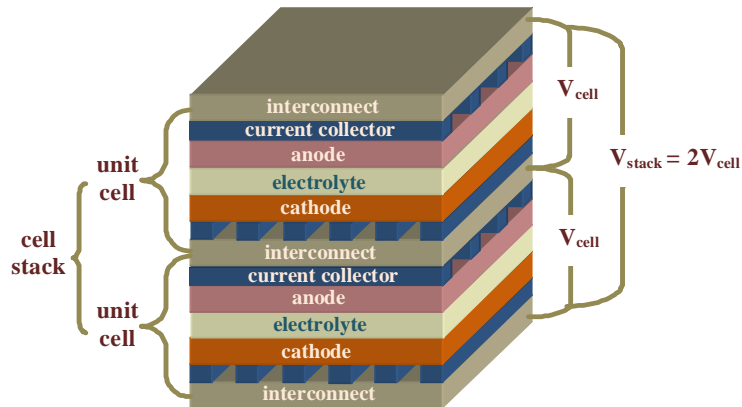
as shown in Table 1. Regardless the specific type of fuel cell, gaseous fuels (usually hydrogen) and oxidants (usually ambient air) are continuously fed to the anode and the cathode, respectively. The gas streams of the reactants do not mix, since they are separated by the gas tight phase of the electrolyte. The electrochemical combustion of hydrogen, and the electrochemical reduction of oxygen, takes place at the surface of the electrodes, the porosities of which are to provide an extensive area for these reactions to be catalysed, as well as to facilitate the mass transport of the reactants/products to/from the electrolyte from/to the gas phase. Under closed circuit, the electrochemical reactions involve a number of sequential steps, including adsorption/desorption, surface diffusion of reactants or products, and the charge transfer to or from the electrode. Charge transfer is restricted to a narrow (almost one-dimensional) three-phase-boundary

(tpb) among the gaseous reactants, the electrolyte, and the electrode-catalyst.

**Table 1** Fuel cell types

	anodic reaction	electrolyte	cathodic reaction
<b>PEFC</b>	$2\text{H}_2 \rightarrow 4\text{H}^+ + 4\text{e}^-$	polymer membranes charge carrier: $\text{H}^+$	$\text{O}_2 + 4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2\text{O}$
<b>SOFC</b>	$2\text{H}_2 + 2\text{O}^{2-} \rightarrow 2\text{H}_2\text{O} + 4\text{e}^-$	mixed ceramic oxides charge carrier: $\text{O}^{2-}$	$\text{O}_2 + 4\text{e}^- \rightarrow 2\text{O}^{2-}$
<b>MCFC</b>	$2\text{H}_2 + 2\text{CO}_3^{2-} \rightarrow 2\text{H}_2\text{O} + 2\text{CO}_2 + 4\text{e}^-$	immobilized molten carbonate charge carrier: $\text{CO}_3^{2-}$	$\text{O}_2 + 2\text{CO}_2 + 4\text{e}^- \rightarrow 2\text{CO}_3^{2-}$
<b>PAFC</b>	$2\text{H}_2 \rightarrow 4\text{H}^+ + 4\text{e}^-$	immobilized liquid $\text{H}_3\text{PO}_4$ charge carrier: $\text{H}^+$	$\text{O}_2 + 2\text{CO}_2 + 4\text{e}^- \rightarrow 2\text{CO}_3^{2-}$
<b>AFC</b>	$2\text{H}_2 + 4\text{OH}^- \rightarrow 4\text{H}_2\text{O} + 4\text{e}^-$	immobilized KOH charge carrier: $\text{OH}^-$	$\text{O}_2 + 2\text{H}_2\text{O} + 4\text{e}^- \rightarrow 4\text{OH}^-$

Besides their catalytic role, electrodes collect (anode) or supply (cathode) the electrons involved in the electrochemical reactions, and should be made of materials with good electrical conductivity. Continuous electrons supply (or removal) is necessary for the electrochemical reactions to proceed, resulting a constant electron flow from the anode to the cathode. At the same time, the electrolyte, by transporting reactants in the form of ionic species, completes the cell circuit. The electro-combustion of hydrogen sustains a difference in the chemical potentials of the electro-active species (conducting ions) between the two electrodes, which is the driving force for the ionic flux through the electrolyte, expressed as the open circuit potential of the cell or its electromotive force (emf).



**Figure 2** Visualization of the physical structure of a planar fuel cell stack

Completing the physical structure of a fuel cell, a current collector, in closed contact to the porous electrodes, facilitates the electrons transport. In actual fuel cell devices, conductive interconnects are used to combine unit cells, in order to upgrade voltage, as illustrated in Figure 2 for the classical planar cell stuck assembly (gaseous fuels and oxidants flow through the current collector formatted channels, in a cross-flow pattern). These interconnects also serve as separator plates between the fuel and the oxidant gaseous streams of successive unit cells, so they must be impermeable to gases. Furthermore, they form the structures for distributing the reactant gases across the electrode surface.

## 2. Operation and Performance

The maximum electrical work ( $W_{el}$ ) of a fuel cell is given by the change in the free energy:

$$W_{el} = \Delta G = -nFE \quad 1.$$

of the overall (combined anodic and cathodic) electrochemical reaction:



where  $n$  is the number of electrons participating the reaction,  $F$  is Faraday's constant (96,487 cb/mole), and  $E$  is the reversible potential of the cell (the open circuit voltage – emf). The difference between  $\Delta G$  and  $\Delta H$  is proportional to the change in entropy ( $\Delta S$ ):

$$\Delta G = \Delta H - T\Delta S \quad 3.$$

where  $\Delta H$  is the total thermal content of the feed and  $T\Delta S$  is the amount of heat produced by a fuel cell operating reversibly. The reversible potential of a fuel cell at temperature  $T$  is calculated from the  $\Delta G$  of the cell reaction, at that temperature:

$$\Delta G = \Delta G^\circ + RT \ln \frac{[C]^c [D]^d}{[A]^a [B]^b} \quad 4.$$

so that the reversible potential, becomes:

$$E = E^\circ + \frac{RT}{nF} \ln \frac{[A]^a [B]^b}{[C]^c [D]^d} \quad 5.$$

the general form of the Nernst equation, where  $\Delta G^\circ$  and  $E^\circ$  refer to 298 K. The ideal performance of a fuel cell is defined by its Nernst potential. Nernst equations, which quantify the relationship between the ideal standard potential ( $E^\circ$ ) and the ideal equilibrium potential ( $E$ ), for the electrochemical reactions of the various types of fuel cells, along the typical values of Nernst potentials, at their operation temperatures are presented in Table 2 [1].

**Table 2** Nernst equations and ideal voltages for the various types of fuel cells

type	overall (anode + cathode) reaction	Nernst equation	T, °C	E, V
AFC	$H_2 + 1/2O_2 + H_2O \rightarrow 2H_2O$	$E = E^\circ + \frac{RT}{2F} \ln \frac{P_{H_2} P_{O_2}^{1/2} P_{H_2O}^{anod}}{P_{H_2O} P_{H_2O}^{cath}}$	100	1.17
PEFC	$H_2 + 1/2O_2 \rightarrow H_2O$	$E = E^\circ + \frac{RT}{2F} \ln \frac{P_{H_2} P_{O_2}^{1/2}}{P_{H_2O}}$	80	1.17
PAFC			205	1.14
SOFC			1100	0.91
MCFC	$H_2 + 1/2O_2 + CO_2 \rightarrow H_2O + CO_2$	$E = E^\circ + \frac{RT}{2F} \ln \frac{P_{H_2} P_{O_2}^{1/2} P_{CO_2}^{anod}}{P_{H_2O} P_{CO_2}^{cath}}$	650	1.03

As noticed from Table 2, and because the entropy change of hydrogen combustion is negative, the reversible potential decreases with temperature by a factor of 0.84 mV/°C (assuming reaction product is liquid water). Furthermore, the actual cell voltage is smaller than its reversible one because of irreversible potential losses (or overpotentials), which originate either from the potential requirements to activate the electrochemical reactions (activation overpotential –  $\eta_{act}$ ), the ohmic losses (ohmic overpotential –  $\eta_{ohm}$ ), or the losses due to the mass transport (gas and electrode's surface diffusion) of the species participating the electrochemical reactions (concentration overpotential –  $\eta_{conc}$ ). Activation overpotential is the primary source of voltage losses at low current densities, expressing, in a sense, the activation energy of the electrochemical reactions to occur, and is described by the Butler-Volmer equation, or its high field approximation, known as the Tafel equation:

$$\eta_{act} = \frac{RT}{\alpha nF} \ln \frac{i}{i_0} \quad 6.$$

( $\alpha$ ,  $i_0$  are the charge transfer coefficient and the exchange current density, expressing the effectiveness of the electrode/electrolyte interface under the specific fuel (anode) or oxidant (cathode) conditions). Ohmic overpotential:

$$\eta_{\text{ohm}} = iR_{\text{ohm}} \quad 7.$$

increases linearly with current (since the resistance of the cell is essentially constant) becomes gradually predominant, as the current density increases. Finally, concentration (mass transport) losses:

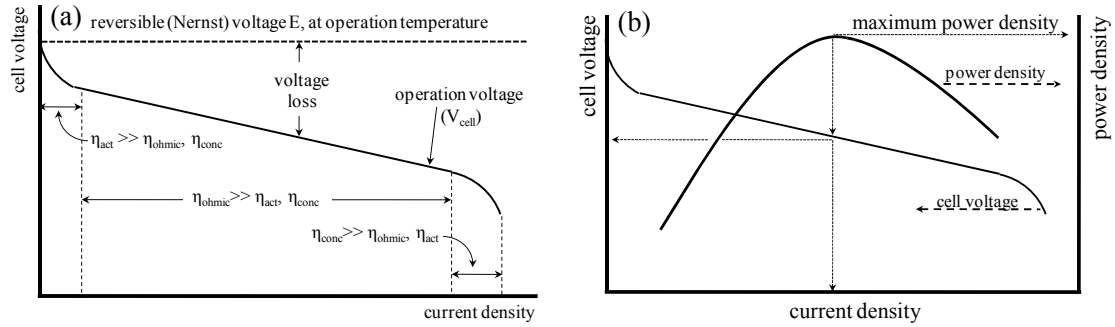
$$\eta_{\text{conc}} = \frac{RT}{nF} \ln \left( 1 - \frac{i}{i_L} \right) \quad 8.$$

( $i_L$  is the limiting or maximum obtainable current due to mass transport) are present over the entire range of current densities, but become prominent at high currents, where it becomes difficult for homogenous or surface diffusion to provide enough electro-active species to the electrode's (or the tpb) reaction sites.

The operational cell voltage is the difference between the potentials of the cathode and the anode (as these potentials are altered due to the corresponding activation and concentration losses of each electrode) minus the ohmic losses, of the various stack components:

$$V_{\text{cell}} = (E_{\text{cath}} - |\eta_{\text{act}}^{\text{cath}}| - |\eta_{\text{conc}}^{\text{cath}}|) - (E_{\text{anod}} + |\eta_{\text{act}}^{\text{anod}}| + |\eta_{\text{conc}}^{\text{anod}}|) - iR \quad 9.$$

Current flow in a fuel cell results in a decrease of cell voltage, revealing the goal to minimize polarization, since the product of  $V_{\text{cell}}$  with the corresponding current density (at each point of the  $I - V_{\text{cell}}$  curve of Figure 2a) gives the specific (per unit of apparent electrode area – power density) electrical power output of the cell. This product tends to be minimized for low and high current densities (when current and operating voltages approach zero, respectively) and exhibits a maximum in between, as shown in Figure 2b.



**Figure 3** Ideal and operational cell voltage (a), and the dependence of power density on cell voltage (b)

The thermal efficiency of fuel cells is defined as the amount of useful energy produced over the consumption of the chemical energy of the fuel (the amount of energy released in the form of heat, during the total combustion of the fuel, known as higher heating value of the fuel). Ideally the electrical work produced in a fuel cell should be equal to the change in Gibbs free energy,  $\Delta G$ , of the overall reaction, and the ideal efficiency for reversible operation at standard conditions, will be:

$$\eta_{\text{ideal}} = \frac{\Delta G^{\circ}}{\Delta H^{\circ}} = \frac{-nFE^{\circ}}{\Delta H^{\circ}} \quad 10.$$

The thermal efficiency of an actual fuel cell, operating irreversibly at temperature  $T$ , reduces to:

$$\eta_{\text{th}} = \frac{-nFV_{\text{cell}}}{\Delta H^{\circ}} = \eta_{\text{ideal}} \frac{-nFV_{\text{cell}}}{-nFE^{\circ}} = \eta_{\text{ideal}} \frac{V_{\text{cell}}}{E^{\circ}} \quad 11.$$

Thus, the efficiency of an actual fuel cell can be expressed in terms of the ratio of the operating cell voltage to the standard cell voltage.

A fuel cell can be operated at different current densities. It seems reasonable to operate the fuel cell at its maximum power density. However, decreasing current density below this value, brings the cell voltage closer to its reversible one, and increases the efficiency. On the other hand, the active cell area must also be increased in order to obtain a given power output, which means that, high efficiencies increase the capital cost, for a certain power level, although it decreases fuel requirements. Balancing between the above, it is

usual practice to operate fuel cells to the left side of its power peak and at a point that yields as a compromise between low operating cost and low capital cost. It is interesting to observe that this situation provides fuel cells with the unique, among other energy conversion technologies, benefit of increased efficiency at part load conditions [1].

For a given fuel cell, it is possible to improve performance by adjusting temperature, pressure, gas composition, reactant utilizations, current density and/or other parameters which influence the ideal cell potential and the magnitude of the voltage losses. The selection of these parameters starts with defining the power level requirements for a specific fuel cell application. Flowingly, the voltage, and current requirements of the fuel cell stack and individual cells need to be determined, at certain operating temperature and (in some cases) pressure. Starting with temperature, its effect on the operational voltage is quite different from its effect on the reversible potential. The latest drops with temperature, while the operating voltage increases, due to the decrease of polarization losses (at higher temperatures the reaction and mass transfer are accelerated, and, in most cases, the ionic conductivity of the electrolyte – the main source of IR losses – increase), resulting in an overall improvement of the performance of the cell. Furthermore, the increase of operating pressure increases the partial pressures of reactants and consequently the reaction and mass transfer rates, improving performance and efficiency. However, pressure increases power needs to compress reactants, and capital costs.

Reactants utilization and gas composition also affect the fuel cell efficiency. Utilization factor ( $U_f$ ) refers to the fraction of the total fuel or oxidant supply, that it is electrochemically consumed. In low-temperature fuel cells (PEFCs, AFCs and PAFCs),  $U_f$  is directly connected to  $H_2$  consumption, which is the only reactant involved in the electrochemical reaction:

$$U_f = \frac{H_{2,in} - H_{2,out}}{H_{2,in}} = \frac{H_{2,consumed}}{H_{2,in}} \quad 13.$$

where  $H_{2,in}$  and  $H_{2,out}$  are the molar flow rates of  $H_2$  at the inlet and the outlet of the fuel cell. Similar is the calculation for the oxidant utilization:

$$U_f = \frac{O_{2,in} - O_{2,out}}{O_{2,in}} = \frac{O_{2,consumed}}{O_{2,in}} \quad 14.$$

for PEMFs, SOFCs and PAFCs. The oxidant utilization in MCFCs and AFCs, where two reactant gases ( $O_2$  and  $CO_2$  in the first and  $O_2$  and  $H_2O$  in the latest) are utilized in the electrochemical reaction, the oxidant utilization is based on the limiting reactant. Frequently  $O_2$ , which is readily available from ambient air, is present in excess, and  $CO_2$  or  $H_2O$  are the limiting reactants. A significant advantage of high-temperature fuel cells such as SOFCs and MCFCs is their ability to use CO as a fuel. The anodic oxidation of CO is slow compared to that of  $H_2$ . However, the water gas shift reaction:



rapidly reaches equilibrium at temperatures over 650 °C, and the equilibrium is shifted to the right because  $H_2O$  and  $CO_2$  are produced, over the anode. Thus,  $H_2$  utilization in high temperature fuel cells is defined as:

$$U_f = \frac{H_{2,in} - H_{2,out}}{H_{2,in} + CO_{in}} = \frac{H_{2,consumed}}{H_{2,in} + CO_{in}} \quad 16.$$

where the  $H_2$  consumed originates both from the  $H_2$  feed and the water gas shift reaction.

### 3. Types of Fuel Cells – Technology Status

A determinative factor concerning the choice of fuel cell type, is hydrogen purity. Low temperature fuel cells require pure hydrogen, because the catalyst exhibits almost no tolerance to sulphur compounds and carbon monoxide, arising problems for hydrogen produced from natural gas. In contrast, SOFCs and MCFCs are more tolerant to impurities, due to their high operating temperatures, and CO, a contaminant for PEM fuel cells, can be used as a fuel. Polymer electrolyte fuel cells (PEFCs), solid oxide fuel cells (SOFCs) and molten carbonate fuel cells (MCFCs) are considered as the most promising candidates for

stationary applications. PEM fuel cells generate power densities up to 4 A/cm<sup>2</sup>, at high efficiencies, while most technologies can hardly reach 1 A/cm<sup>2</sup>. This characteristic, in combination to weight, volume, and cost advantages, makes this type of fuel cells the most attractive for both mobile and stationary applications. SOFCs and MCFCs appear to have similar prospects to gain a part of the market in the coming decades [1,2].

### 3.1 Polymer Electrolyte Fuel Cells

A typical PEM fuel cell assembly includes the polymeric proton exchange membrane, on the opposite sides of which two porous electrocatalytic layers (electrodes) are supported. Two conductive and porous collectors are layered over the electrodes in closed contact to the hard-plate interconnects, which form the reactants and products flow channels [1,2]. The proton exchange membrane consists of perfluorosulfonic acid polymers. These materials are gas-tight electrical insulators, in which the ionic transport is highly dependent on the bound and free water in the polymer structure. Nafion of the perfluorosulfonic acid family is the most commonly used material. Its Teflon-like structure is bonded to perfluorinated side chains with terminal sulfonic acid groups (SO<sup>3-</sup>). These groups are hydrophilic, and the degree of their hydration is determinative for the ionic conductivity, the gas permeability, and the elasticity [1]. Nafion membranes exhibit high thermal stability and chemical durability against Cl<sub>2</sub>, H<sub>2</sub>, and O<sub>2</sub> attacks at temperatures up to 125°C [1,4,5], and their operational lifetime has been proved for over 50,000 hours. Apart Nafion, research has also focused on polybenzimidazole (PBI) electrolytes [2,6], which can operate at temperatures over 160°C, and annihilate CO poisoning [7,8].

The electrode-catalyst layer, for both the anode and the cathode, is in intimate contact with the membrane and consists of micro-dispersed platinum in a binder. The degree of intimacy between its particles and the membrane is crucial for the optimal proton mobility. The binder stabilizes the catalytic particles within the electrode structure and could be either hydrophobic (usually polytetrafluoroethylene) or hydrophilic (usually perfluorosulfonic acid). Platinum loading, an important cost factor for PEFCs, has decreased to 1.0 mg Pt/ cm<sup>2</sup> of membrane (total on anode and cathode) – from 2.0 – 4.0 mg Pt/cm<sup>2</sup> [1].

The Nafion membrane is sandwiched between two porous and conductive carbon-based cloths, which support the membrane, diffuse the gaseous reactants and products and collect or supply the electrons. This layer incorporates a hydrophobic material (usually polytetrafluoroethylene) to prevent withholding water within its pores, and to facilitate the removal of product water in the cathode. The current collecting cloth is in closed contact (the membrane-electrode-cloth assembly is sandwiched between) with interconnecting carbon composite plates, for current collection, gas tightness, gas distribution, and thermal management. Flow paths for reactants, products and/or the cooler are printed on either side of these plates. In most PEFCs cooling is accomplished by circulating water that is pumped through integrated coolers within the stack, so that the temperature gradient across the cell is kept to less than 10°C.

Because of Nafion membranes, PEFCs operate at temperatures typically not higher than 60 – 80°C. At these temperatures CO is strongly chemisorbed on platinum, poisoning its catalytic activity and reducing the performance of the cell. The effect is reversible for only up to 50 ppm CO, while reformed and shifted hydrocarbons contain over 10,000 ppm CO. Although electrolysis H<sub>2</sub> is favored for PEFC applications, in combined reformer – PEFC systems these concentrations can be eliminated by preferential oxidation (a process that selectively oxidizes CO in rich H<sub>2</sub> streams, over precious metal catalyst). Recently PEFC research has focused on upgrading temperatures over 160°C using polybenzimidazole (PBI) electrolytes [1,2,6]. At these temperatures not only CO poisoning is eliminated, but also and because PBI requires lower water content to operate, water management is simplified [7,8].

As a proton is conducted through the membrane, it drags 1 – 2.5 water molecules with it [1,9], thus it is critical to maintain the water content of the electrolyte (the conductivity of the electrolyte is maximised when the membrane is fully saturated). On the other hand, and due to operation at less than 100°C at atmospheric pressure, liquid water is produced at the cathode. Thus, in case the anode is drier than the cathode, the back-diffusion of water from the cathode to the anode inhibits the protonic flow [9,10]. Furthermore, in case of water excess, the electrodes will be flooded blocking the pathways of reactants (products) to (from) the electrode/electrolyte interface, while in case of water shortage, the membrane will be dehydrated and destroyed [11]. Despite the complexity, effective forms of water management have been developed based on continuous flow fields and appropriate operating adjustments focusing on controlled humidification of the anode gas in case of water shortage and temperature rise in case of flooding [12,13].

With operation voltages of 0.7 – 0.75 V, the maximum efficiency of PEFCs can be as high as 64%. In today's applications, certain losses and ancillary equipment lower the efficiency, resulting in a situation in

which PEFCs are more efficient than internal combustion engines only for operation at partial loads [2,14]. Current – Voltage performance characteristics of up to 5 kWe PEMFC are in the range of 0.5 – 0.76 V/cell for current densities of 0.55 – 1 A/cm<sup>2</sup> and power densities of 0.22 – 0.57 W<sub>e</sub>/cm<sup>2</sup> [15]. Operating temperature has a significant influence on performance (decrease of the ohmic resistance of the electrolyte and mass transport limitations), resulting a voltage gain of 1.1 - 2.5 mV / °C [1,16]. Improving the cell performance through temperature, however, is limited by water management issues. The goal for stationary PEFC operating life is 40,000 – 60,000 hours or 5 – 8 years [17]. This life depends to a large extent on the operating conditions, such as the external temperature at start-up, excessive or insufficient humidification, and fuel purity. The principal areas of development concern improved cell membranes and electrode designs, targeting to improve performance and reduce cost [1,18,19,20].

### 3.2 Solid Oxide Fuel Cells

Zirconia, stabilized with 8 – 9 % yttria (Yttria Stabilized Zirconia – YSZ) is the most commonly used electrolyte for SOFCs because it exhibits predominant ionic conductivity (O<sup>2-</sup> transport number close to unity) over a wide range of oxygen partial pressures (1 to 10<sup>-20</sup> atmospheres). YSZ provides sufficient conductivity at very high temperatures (900 – 1000 °C), and requires expensive high temperature alloys to house the fuel cell, increasing the cost substantially. These costs could be reduced if the operating temperatures were lowered to between 600 – 800 °C, allowing the use of less expensive structural materials such as stainless steel. To lower the operating temperature, either the conductivity of the YSZ must be improved by producing thinner electrolytes, or alternative electrolytic materials must be developed. To reduce the resistivity of the electrolyte, development has focused on reducing electrolyte thickness from 150 μm to less than 20 μm. Wang, et al. fabricated thin-film YSZ electrolytes between 3 and 10 μm. Furthermore, scandium doped zirconia (SDZ) is more conductive than YSZ, permitting the reduction of the operating temperature by 50 – 100 °C, while gadolinium doped ceria (GDC) is even more conductive, but above 600°C it is partially reduced in hydrogen, and the developed electronic conductivity short-circuits the cell. Lanthanum gallate with strontium doping on the A-site of the perovskite structure and magnesium on the B-site could be used at temperatures as low as 600 °C, but matching the thermal expansion coefficients, mechanical strength, and chemical compatibility need further development [1,20].

SOFC anodes are fabricated from composite powdered mixtures of ceramic electrolyte materials – YSZ, GDC, or SDC – and nickel oxide (cermets). The nickel oxide is reduced to nickel metal prior to operation. The electrolyte skeleton inhibits sintering of the metal particles and provides comparable to YSZ thermal expansion coefficient. Typical anode materials have nickel contents of approximately 40 % volume, after nickel reduction. The anode structure is fabricated with a porosity of 20 – 40% to facilitate mass transport of reactant and product gases, and 5 – 20 m<sup>2</sup>/g surface areas. Concerning cathode, lanthanum strontium manganite (LSM) perovskite is the most frequently used material, offering excellent thermal expansion match with YSZ and providing good performance above 800°C. For lower temperatures, a range of alternative perovskites are available (lanthanum strontium ferrite – LSF, lanthanum strontium cobalt ferrite – LSCF, lanthanum strontium manganese ferrite – LSMF and others). Improved performance of the cathode can be obtained by blending it with electrolyte material, typically at 40 % volume.

Interconnects (usually doped lanthanum chromite), must be chemically and dimensionally stable in both oxidizing and reducing conditions. They must have similar, to the rest of components, thermal expansion coefficients. One of the most significant changes resulting from operation below 800°C is the shift from ceramic to metallic interconnects, which will reduce the cost of the stack. The seal of the cell must also have similar thermal expansion coefficient, along with chemical compatibility with the rest of the stack components and the gaseous constituents of the highly oxidizing and reducing environments of the cathode and the anode respectively. In addition, the seal should be electrically insulating to prevent shorting within the stack. A possible alternative to glass is the use of compressive, non-bonding seals, and the use of micas, or of silicate platelets [1,21,22].

Planar SOFCs are composed of flat, ultra thin ceramic plates, which allow them to operate at 800°C or even less, and enable less exotic construction materials. P-SOFCs can be either electrode or electrolyte – supported. Electrolyte – supported cells use YSZ membranes of about 100 μm thickness, the ohmic contribution of which, is still high for operation below 900°C. In electrode – supported cells, the supporting component can either be the anode or the cathode. In these designs, the electrolyte is typically between 5 – 30 μm, while the electrode thickness can be between 250 μm – 2 mm. In the cathode – supported design, the YSZ electrolyte and the LSM coefficients of thermal expansion are well matched, placing no restrictions on electrolyte thickness. In anode – supported cells, the thermal expansion coefficient of Ni –

YSZ cermets is greater than that of the YSZ electrolyte. This limits the electrolyte thickness at about 30  $\mu\text{m}$ . Nevertheless, in anode – supported cells concentration polarization is much lower since the effective diffusivity of the fuel is typically 3 to 4 times greater than that of air. Anode-supported cells, and lower operating temperatures create the potential for lower cost stainless steel interconnects, which are protected against oxidation by surface films of chromium oxide, aluminum oxide, or some other stable oxides [1].

Tubular SOFCs has the advantage not to require extensive gas sealing, which enables operation at higher temperatures. The closed at one end, cathode supporting tube, is fabricated by extrusion and sintering and has a porosity of 30 – 40 %. The interconnect (doped lanthanum chromite) is applied to the cathode tube as a narrow strip prior to depositing the electrolyte by masking the rest of the tube. Similarly, the interconnect strip is masked when the electrolyte is applied. Thin electrolyte structures of about 40  $\mu\text{m}$  thickness can be fabricated by EVD, tape casting or other ceramic processing techniques. The anode is subsequently formed on the electrolyte by slurry deposition.

The operation voltage of a SOFC is linearly reduced by current density, by an indicating coefficient of 0.73 mV/mA/cm<sup>2</sup>, at 1000 °C and for usual component materials and thicknesses. Voltage losses in SOFCs are primarily governed by ohmic losses (45, 18, 12 and 25% from the cathode, the anode, the electrolyte, and the interconnect). The voltage loss is also a strong function of temperature. The performance improves with the use of O<sub>2</sub> rather than air as oxidant, which suggests that concentration polarization is significant during cathodic O<sub>2</sub> reduction in air. Although, both H<sub>2</sub> and CO can be regarded as fuels, the theoretical potential for the H<sub>2</sub> exceeds that for CO at temperatures above 800 °C. Consequently, increased H<sub>2</sub> in the fuel gas yields higher open circuit potentials and higher efficiencies. Furthermore, low concentrations of H<sub>2</sub> and/or CO increase concentration polarization, and thus cell voltage decreases with fuel utilization. Fuel and oxidant utilizations are usually of the order of 85 and 25 %, respectively. Typical operation characteristics of currently operating small scale (up to 5 kW<sub>e</sub>) SOFCs are ranged between 0.6 – 0.79 V, at 0.2 – 0.8 A/cm<sup>2</sup>, and 750 – 800 °C, resulting power densities of the order of 0.16 – 0.55 W/cm<sup>2</sup>. When combined with gas turbines, SOFCs are expected to achieve up to 60 – 70 % electrical efficiency and up to 80 – 85 % cogeneration efficiency. The durability of SOFCs depends upon the type of design and the operating conditions. Laboratory or pilot tests have demonstrated lifetimes of up to 8 years, at steady-state conditions, but just 50 on/off cycles can cause irreversible damages due to thermal stresses [1,23-26].

### 3.3 Molten Carbonate Fuel Cells

MCFCs can operate efficiently with CO<sub>2</sub> containing fuels, i.e. hydrocarbon reformates, coal and biomass derived gases, although the need for CO<sub>2</sub> at the cathode requires, either its transfer from the anode exit (usual practice) or its production by the combustion of the anode exhaust gas. The electrolyte of MCFCs is a combination of alkali carbonates, retained in a ceramic matrix of LiAlO<sub>2</sub>. The electrolyte matrix is  $\alpha$  or  $\gamma$ -LiAlO<sub>2</sub>. In long-term operation, significant particle growth, and  $\alpha$  to  $\gamma$  phase transformation, lead to detrimental changes in the pore structure of the matrix, especially in low CO<sub>2</sub> atmospheres, while thermal cycling below the carbonate's freezing temperature can induce cracking of the matrix. MCFCs operate at 600 – 700 °C, where these carbonates form a highly CO<sub>3</sub><sup>2-</sup> conductive molten salt [1,27]. The electrolyte composition affects the performance and endurance of MCFCs, and it is responsible for over 70 % of the cell's ohmic losses. Li<sub>2</sub>CO<sub>3</sub> exhibit higher ionic conductivity than Na<sub>2</sub>CO<sub>3</sub> and K<sub>2</sub>CO<sub>3</sub>, however, its gas solubility and diffusivity are lower. Present electrolytes are lithium potassium carbonates (Li<sub>2</sub>CO<sub>3</sub>/K<sub>2</sub>CO<sub>3</sub> - 62:38 mol %) for atmospheric pressure operation and lithium sodium carbonates (Li<sub>2</sub>CO<sub>3</sub>/Na<sub>2</sub>CO<sub>3</sub> - 52:48 to 60:40 mol%), for operation at higher pressures. Electrolyte management relies on a balance of capillary pressures, which permits the matrix and the porous electrodes to remain completely and partially filled with molten carbonate. The electrolyte has a low vapor pressure, at the operating temperatures, and slowly evaporates. An area for electrolyte improvement is to prevent gas crossover from one electrode to the other [1].

Ni state of the art anodes contain Cr to eliminate the problem of sintering. However, Ni-Cr anodes are susceptible to creep, while Cr can be lithiated by the electrolyte and consume carbonate, leading to efforts for Cr decrease. State of the art cathodes are made of lithiated NiO. Dissolution of the cathode is probably the primary life-limiting constraint of MCFCs, particularly under pressurized operation. For atmospheric cells, the use of more basic melts (such as Li/NaCO<sub>3</sub>) reduces dissolution. To operate at higher pressures and CO<sub>2</sub> contents, alternative cathodes materials and electrolyte additives are examined. The major considerations with Ni-based anodes and NiO cathodes is structural stability and NiO dissolution, which become worse at high CO<sub>2</sub> partial pressures. Nevertheless, Ni dissolution and subsequent precipitation is not an issue for the desired 40,000 – hour lifetimes at atmospheric pressure, while at 10 atm only about



5,000 to 10,000 hours lifetimes can be attained [1,28].

The present bipolar plate consists of the separator, the current collectors, and the seal. The bipolar plates are usually fabricated from thin sheets of a stainless steel alloy coated on one side by a Ni layer, which is stable in the reducing environment of the anode. On the cathode side, contact electrical resistance increases as an oxide layer builds up.

The high operating temperature is needed to achieve sufficient electrolyte conductivity. Most MCFC stacks operate at 650 °C, as a compromise between high performance and stack life, because, over 650 °C there are increased electrolyte losses due to evaporation and increased material corrosion. The influence of temperature on the reversible potential depends on several factors, one of which involves the equilibrium composition of the fuel gas. The water gas shift reaction achieves rapid equilibrium at the anode and CO serves as an indirect source of H<sub>2</sub>. The voltage of MCFCs varies with the composition of the reactant gases. Increasing the reactant gas utilization generally decreases cell performance. A compromise leads to typical utilizations of 75 to 85% of the fuel. The electrochemical reaction at the cathode involves the consumption of two moles CO<sub>2</sub> per mole O<sub>2</sub>, and this ratio provides the optimum cathode performance.

Gasified coal (or biomass) is expected to be the major source of fuel gas for MCFCs, but fuels derived from these sources contain a considerable number of contaminants. Sulfur compounds in low ppm concentrations are detrimental to MCFCs. At atmospheric pressure and high gas utilization (~75%), <10 ppm H<sub>2</sub>S in the fuel can be tolerated at the anode, and <1 ppm SO<sub>2</sub> is acceptable in the cathode. These concentration limits increase with temperature, and decrease with pressure. Halogen-containing compounds are also destructive because they can lead to severe corrosion of cathode hardware, while NO<sub>x</sub>, produced by combustion of the anode effluent, can react irreversibly with the electrolyte in the cathode compartment to form nitrate salts.

Endurance is a critical issue in the commercialization of MCFCs. Adequate cell performance must maintain an average potential degradation no greater than 15 mV/a over a cell stack lifetime of 5 years, while state-of-the-art MCFCs exhibit an average degradation of 40 mV/a. At full load, MCFC system can achieve efficiencies up to 55%, which drops at partial loads. Typical MCFCs operate in the range 100 – 200 mA/cm<sup>2</sup>, at 750 – 900 mV/cell, achieving power densities even above 150 mW/cm<sup>2</sup> [1,2].

### 3.4 Phosphoric Acid Fuel Cells

PAFCs were the first fuel cell technology to be commercialized and represented almost 40 % of the installed fuel cell units in 2004 [29]. Most of the demonstration units are in the range of 50 – 200 kW, but larger plants (1 – 10 MW) or smaller systems (1 – 10 kW) have also been built [30-33]. Lifetimes of 5 years with > 95 % durability have been demonstrated [34]. Phosphoric acid electrode/electrolyte technology has reached of maturity. However, further increases in power density and reduced cost are needed to achieve economic competitiveness [1].

Concentrated (up to 100%) H<sub>3</sub>PO<sub>4</sub> in silicon carbide matrix, is the proton conducting electrolyte. The relative stability of concentrated H<sub>3</sub>PO<sub>4</sub> is high, enabling PAFCs to operate at temperatures up to 220°C. Common systems operate between 150 and 220°C, because at lower temperatures, H<sub>3</sub>PO<sub>4</sub> is a poor ionic conductor, and CO poisoning of the Pt anodic electrocatalyst is severe. The electrolyte vaporizes and a portion of H<sub>3</sub>PO<sub>4</sub> escapes in the air stream. The use of concentrated acid minimizes the water vapor pressure and water management becomes easy. An electrolyte reservoir plate, made of porous graphite, can also be used to replenish H<sub>3</sub>PO<sub>4</sub>, towards achieve a goal of 5 - year cell lifetime.

Pt supported on carbon black is the state of art electrocatalyst, with Pt loadings of about 0.1 mg/cm<sup>2</sup> in the anode and 0.50 mg Pt/cm<sup>2</sup> in the cathode. Transition metal (e.g., iron, cobalt) organic macrocycles (tetramethoxyphenylporphyrins, phthalocyanines, tetraazaannulenes and tetraphenylporphyrins) and Pt alloys with transition metals (Ti, Cr, V, Zr), have been evaluated as cathodic electrocatalysts. The electrodes contain a mixture of electrocatalyst and a 30 – 50 wt% PTFE polymeric binder. The PTFE binds the carbon black particles together to form a porous structure, which is supported on a porous graphite substrate. The graphite substrate, serves as a support for the electrocatalyst, and as the current collector. Bipolar plates separate the individual cells and connect them electrically. They are made of graphite resin mixtures, carbonized at 2,700 °C, which are corrosion stable in PAFC operating environments. The bipolar plates form gas channels that feed the reactant gases to the porous electrodes and remove the products. Graphite bipolar plates are sufficiently corrosion-resistant for lifetimes of 40,000 hours. Generated heat during is removed by either liquid (two-phase water or a dielectric fluid) or air coolants that are routed through cooling channels located every fifth cell in the stack.

One of the major causes of degradation is the electrode flooding and drying, by the migration of  $H_3PO_4$  between the matrix and the electrodes, during cell load cycling. Optimized wettability and heat treatment of the cathodic carbon support can improve cell life performance while keeping power density high. Heat treating of carbon supports around 3,000 °C, better cell temperature control, increased  $H_3PO_4$  inventory, can further help towards 40,000 cell life.

CO absorption affects Pt electrode catalysts. Both temperature and CO concentration have a major influence on this effect.  $H_2S$  and COS reduce the effectiveness of the catalysts. The concentration levels of total sulfur ( $H_2S + COS$ ) are 50 ppm. The effect of  $H_2S$ , however, decreases with increasing cell temperature. The fuel stream must contain 1 vol % or less of CO, less than 50 ppmv of COS plus  $H_2S$ , and less than 20 ppm of  $H_2S$ . Especially  $NH_3$ , in the fuel or oxidant gases, reacts with  $H_3PO_4$  to  $(NH_4)H_2PO_4$ , which decreases the rate of  $O_2$  reduction. Consequently, the amount of nitrogen must be limited to 4% because it will react with hydrogen to form  $NH_3$ .

Pressure increase enhances the performance of PAFCs, due to lower diffusion polarization at the cathode and an increase in the reversible cell potential. The increase in temperature also has a beneficial effect on performance (0.55 – 1.15 mV/°C voltage gain), because activation, mass transfer, and ohmic losses are reduced, while the kinetics of oxygen reduction improves. Furthermore, temperature enhances anode tolerance to CO. On the other hand, increasing utilization or decreasing inlet concentration results in decreased cell performance, due to concentration polarization and Nernst losses. State-of-the-art utilizations are on the order of 85% and 50% for the fuel and oxidant, respectively.

The voltage that can be obtained from a PAFC is reduced by ohmic, activation, and concentration losses, which increase with current density by a factor of 0.45 – 0.75 mV/mA/cm<sup>2</sup>, for atmospheric, and 0.4 – 0.5 mV/mA/cm<sup>2</sup>, for high pressure (up to 8 bar) operation. Most of the polarization occurs at the cathode, and it is greater with air (560 mV at 300 mA/cm<sup>2</sup>) than with pure oxygen (480 mV at 300 mA/cm<sup>2</sup>). The anode polarization is very low (-4 mV/100 mA/cm<sup>2</sup>) with pure  $H_2$ . The ohmic loss is also relatively small, amounting to about 12 mV at 100 mA/cm<sup>2</sup>. Typically, PAFCs operate in the range of 100 – 450 mA/cm<sup>2</sup> at 600 – 800 mV/cell, achieving power densities of 100 – 330 mW/cm<sup>2</sup>, and electrical efficiencies of 40 – 55 % HHV [1,35]. Voltage and power constraints arise from increased corrosion of platinum and carbon components at cell potentials above approximately 800 mV.

One of the primary areas of research is in extending cell life. State-of-the-art PAFCs exhibit degradation rates of about 15 – 25 mV/a [1]. The goal is to maintain the performance of the cell stack at 40,000 hours. UTC Fuel Cells has demonstrated over 25 atmospheric pressure PAFC plants of 200 kW, that have operated for 4 – 5 years [1]. The performance degradation rate was less than 2 – 4 mV/1,000 hours. UTC Fuel Cells reported a similar degradation rate in 2002 for power units operating up to 40,000 hours. The efficiency of latest power plants at the beginning of life is 40% LHV. Initially, efficiency reduces quickly to 38%, but then there is a small decrease in efficiency over the next 40,000 hours resulting in an average efficiency over life of 37%.

### 3.5 Alkaline Fuel Cells

AFC were the first fuel cell technology, which was demonstrated in a practical application – i.e. the powering NASA space missions since 1960s [1]. Fast reaction kinetics, leading to higher cell voltages and system efficiencies, is commonly stated as the main advantage of AFCs [36]. However, this advantage is eliminated when AFCs are fuelled by natural gas [35]. The electrolyte is concentrated (85 wt%) KOH in high temperature AFCs (~250°C), or less concentrated KOH (35 – 50 wt%), in low temperature AFCs (<120°C), and it is retained in an asbestos matrix. CO is a poison, and  $CO_2$  reacts with KOH to  $K_2CO_3$ , so that even the small amounts of ambient  $CO_2$  are detrimental. Along with restricted lifetimes (due mostly to the presence of  $CO_2$  traces), the cost of  $CO_2$  scrubbing in both the air and the fuel, can explain the limited number of companies that persist in AFCs, focusing primarily to niche applications [1,35,37,38]. Nevertheless, cost projections for AFCs can present a more optimistic picture, in case evolution of the basic materials and construction techniques results in lower manufacturing costs [35,39,40].

The electrolyte is KOH water solutions with concentrations of 6 – 12 N. The 35% KOH electrolyte (low temperature AFCs) is usually replenished by a reservoir at the anode side. The use of the less expensive NaOH has also been considered, but minimal cost advantages are outweighed by the lower conductivity. However, NaOH increases the lifetime, because sodium carbonate (gradually formed in  $CO_2$  presence), forms smaller crystals, which do not harm the carbon pores of the electrodes. The fuel/oxidant separator (electrolyte matrix) of AFCs is usually asbestos, but potassium titanate, ceria, or zirconium phosphate gel matrixes, are also used. In most cases noble metal at high loadings (i.e. 80% Pt – 20 % Pd),

are used as electrocatalysts, although non-noble metals (high surface area Raney nickel anodes and Raney silver cathodes containing small amounts of Ni, Bi, and Ti), or even spinels and perovskites are being considered in several attempts to lower the cost. Noble metals loadings are of the order of 5g/m<sup>2</sup>. The electrocatalyst is dispersed on carbon based porous electrodes to ensure gas diffusion. The typical arrangement includes nickel current collectors and gold-plated magnesium is bipolar plates [1].

Non-hydrocarbon, pure H<sub>2</sub> or cracked ammonia (25% N<sub>2</sub>, 75% H<sub>2</sub>, and residual NH<sub>3</sub>) can be fed directly to the anode. Due to the high diffusion rate of hydrogen, in case of NH<sub>3</sub> feed, result only a very small decrease in cell's potential, at medium current densities. Gas purification is necessary for H<sub>2</sub> from carbon-containing sources.

Multistep physical separation or chemical separation methods are mostly used in order to minimize CO<sub>2</sub> in the fuel and the oxidant streams. Cryogenic separation (CO<sub>2</sub> condensation, low temperature distillation or CO<sub>2</sub> liquefaction) can also be used, in cases of liquid or high purity H<sub>2</sub> supply. Membrane (polymer or palladium/silver) separation is also a promising CO<sub>2</sub> removal option. In this case, the membrane transports H<sub>2</sub> by diffusion. High cost, low H<sub>2</sub> fluxes and membrane degradation, along with the necessity for high pressure gradients, across the membrane, are the main drawbacks of these techniques.

The typical performance of this AFC cell is in the range 0.65 – 0.85 V, at 100 – 250 mA/cm<sup>2</sup>, and 60 – 180 mW/cm<sup>2</sup> – UTC Fuel Cells has demonstrated 3.4 W/cm<sup>2</sup> at 0.8 V and 4.3 A/cm<sup>2</sup> [35,39,41]. AFC have failed to reach commercialization so far due to problems related to lifetime and CO<sub>2</sub> degradation [39,40]. Degradation ranges between 160 – 200 mV/a, and lifetimes of 4 – 10 thousand hours (half to one year) have been demonstrated [35,41]. However, the average stack lifetime does not exceed 4,000 hours [1,42]. For large scale applications, economics demand operating times exceeding 40,000 hours, imposing the most significant obstacle to AFCs commercialization [1].

## 4. Fuel Cell Cost Considerations and Market Development

As a potential new product, the cost of ownership and operation will be critical for fuel cells commercialization. This, total cost can be split down to fuel and other operating costs and the initial capital cost. The main component of the initial cost is the manufacturing cost, which is strongly related to the production volume and the incorporation of economies of scale.

### 4.1 Projected and Allowable Fuel Cell Cost

The manufacturing cost of a PEM fuel cell stack includes the individual costs of the membrane, the electrodes, the platinum catalyst, the bipolar plates, the peripheral materials and a minor share of the assembly costs. An overview of the 2004 PEFC cost estimates is presented in Table 3. The total manufacturing cost of approximately 1,300 €/kW<sub>el</sub><sup>1</sup> was dominated by the cost of bipolar plates and electrodes, both manually manufactured at the moment, while their future large-scale production is expected to lead to significant cost decrease [43].

**Table 3** Estimates of current PEM costs [43]

	€/m <sup>2</sup>	€/kW <sub>e</sub>	share, %
membrane	357	179	14
electrodes	1016	509	39
bipolar plates	1179	589	45
platinum	34	17	1
peripherals	11	6	0
assembly		6	0
total		1304	100

The main differences in high temperature fuel cell stack cost structure, relate to the fact that they do not contain high-cost precious metals, on one hand, and that they demand more complex manufacturing process, on the other. It must be noticed therefore, that, the fuel cell stack is, in many cases, responsible for less than the one third of the total capital cost of a fuel cell system [44], and that a large portion of the total cost is caused by fuel pretreatment (reforming, cleaning etc), plant control, and power conditioning. For

<sup>1</sup> In all cases required, the exchange rate was 1 € = 1.4 \$.

small-scale SOFC systems, the cost of the stack is of the order of 40 – 45 % of the total cost, with the rest 55 – 60 % referring to the balance of the system. This cost structure could very well be valid for PEMFC systems, although for high temperature stacks, insulation can be an important factor, especially for low nominal power outputs [1]. Small systems of few kW<sub>e</sub> are not likely to operate under high pressure. While this simplifies the design and reduces cost for compressors and expanders it might also have a negative effect on the power density. Currently available high temperature fuel cell systems reach electrical capacities of around 250 kW<sub>e</sub> (for these systems, the integration of a gas turbine can raise electric efficiency up to 60%). Table 4 indicates the current investment costs for stationary high temperature fuel cells [2,45]. Taking into account those costs, along with the, difficult to estimate, profit margins, and the quite high R&D costs related to the present fuel cell production, the retail prices of smaller systems (up to 5 kW) range between 10.000 and 50.000 €/kW<sub>el</sub>, while larger ones are estimated between 5.000 and 18.000 €/kW<sub>e</sub> [3]. On the other hand, the manufacturing cost of PEM fuel cells is reported to vary depending on scale, power electronics and reformer requirements, with retail prices varying between \$3,000/kW and \$6,000/kW [46-48].

**Table 4** Estimates of current SOFC and MCFC distributed power generation system cost [45]

	SOFC		MCFC	
	€/kW <sub>e</sub>	share, %	€/kW <sub>e</sub>	share, %
fuel cell stack	4714	42	4661	50
boiler	4672	41	2146	23
operating system	1231	11	820	9
reformer	52	0	544	6
heat exchanger	274	2	286	3
burner	109	1	258	3
air supply	118	1	31	0
inverter	151	1	88	1
frame	0	0	500	5
Total	11319		9334	

#### Technology Advancements and Learning Effects

Whether fuel cell components are manually manufactured, or produced by large-scale industrial processes, is widely recognized as a point of major importance, in order to reduce the fuel cell costs and retail prices. In 2000, the estimated production cost was around 100 €/kW<sub>e</sub> for an annual production 500,000 PEMs, optimized for power output (which are expected to have significantly lower costs per kW<sub>e</sub> due to higher power densities) [49]. In 2005, Ballard claimed that PEM manufacturing cost could be reduced to 75 €/kW<sub>e</sub>, even with today's technologies, materials and processes, for the aforementioned annual production rates, aiming to 20 €/kW<sub>e</sub> by 2010 [2,50].

**Table 5** Estimates of future PEM stack costs at a cumulative production of 250,000 MW<sub>e</sub>/a [43]

	€/m <sup>2</sup>	€/kW	share, %
membrane	36	9 – 12	16 – 25
electrodes	69 – 107	17 – 36	48 – 49
bipolar plates	25 – 65	6 – 21	17 – 29
platinum (catalyst)	6	1 – 2	3 – 4
peripherals	3	1	1 – 2
assembly		1	2 – 4
total		36 – 74	100

The key factors to reduce the cost of PEM fuel cells from around 1,300 \$/kW to less than 100 €/kW are [2,3]:

- the mass-production of membranes, electrodes and bipolar plates and the possible use of new materials
- the production increase to 100,000 m<sup>2</sup> per year
- the increase in power density from 2 kW<sub>e</sub>/m<sup>2</sup> to 3 or even 5 kW<sub>e</sub>/m<sup>2</sup> and
- the achievement of higher stack lifetimes

Current PEM fuel cells use nafion membranes, of 50 to 175  $\mu\text{m}$  typical thickness, with an estimated cost of up to 450  $\text{€}/\text{m}^2$ , which corresponds to 110 – 250  $\text{€}/\text{kW}_e$ . Membranes are quite possible to undergo substantial technical and economic breakthroughs, within the decade (e.g. the use of alternative materials like the organically modified silicates), which may result a 10 – 20 fold cost reduction [2].

The cost of electrodes also depends on production technologies, materials and volumes. Automated production at large scales may lower the corresponding costs to even 100  $\text{€}/\text{m}^2$  [2]. Current systems, operating at 80  $^\circ\text{C}$ , need about 5  $\text{g}/\text{m}^2$  of platinum for both the anode and cathode, or 10  $\text{g}/\text{m}^2$ , in total (5  $\text{g Pt}/\text{kW}_e$  for power densities of about 2  $\text{kW}_e/\text{m}^2$ ). New membranes, operating above 100  $^\circ\text{C}$ , are expected reduce required platinum towards an estimated goal of 0.2  $\text{g}/\text{kW}_e$ . Platinum loading of the anode can easily be reduced without affecting the performance. However, reducing platinum at the cathode to 2 – 4  $\text{g}/\text{m}^2$ , within the current catalyst systems, results in efficiency losses of 2 – 4 % [14]. Improved diffusion media and electrode structures can increase the power density, and reduce the platinum load accordingly. New electrode production technologies, leading to larger Pt surface areas, can also reduce the platinum needs, while new more active Pt cobalt and chromium alloys seem capable of even a threefold activity increase [51]. A possible barrier to the full market expansion for PEM fuel cells could lie in the potential global production capacity of platinum, which today is of the order of 200 t/a, and even platinum recycling or the use of other precious metals (palladium, ruthenium) may not be enough to meet demand. Thus, new active catalysts or high-temperature membranes – that do not use Pt – are critical not only for lowering costs but also for securing PEM commercialization at full potential.

**Table 6** Estimations for 2020's SOFC and MCFC system cost [45]

	SOFC (200 kW)		MCFC (300 kW)	
	$\text{€}/\text{kW}_e$	share, %	$\text{€}/\text{kW}_e$	share, %
fuel cell stack	396	33	418	35
boiler	382	32	311	26
operating system	104	9	119	10
reformer	52	4	44	4
heat exchanger	66	6	60	5
burner	38	3	47	4
air supply	38	3	9	1
inverter	66	6	69	6
frame	42	4	101	9
<b>Total</b>	<b>1184</b>		<b>1179</b>	

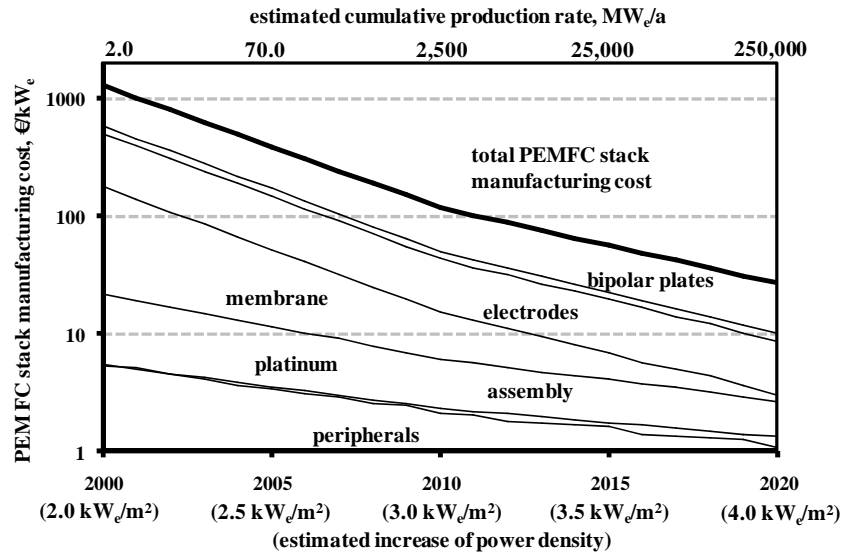
Bipolar plates are currently made from milled graphite or gold-coated stainless steel. Ongoing research is aiming to replace these materials with polymers or low-cost steel alloys, which will allow the use of low-cost production techniques. Even today, bipolar plates can be produced at 200  $\text{€}/\text{kW}$ , if the production volume increase to 10,000 units/a, and even below 20  $\text{€}/\text{kW}$  for 1 million/a, i.e. 10 – 30  $\text{€}/\text{kW}_e$  for power densities of 2 – 6  $\text{kW}_e/\text{m}^2$  [2].

**Table 7** Comparison of conventional and SOFC CHP systems [2]

	conventional	SOFC 2010	SOFC 2030
specific investment, $\text{€}/\text{kW}$	1000	5000	1000
electrical power, $\text{kW}_e$	200	200	200
thermal power, $\text{kW}_t$	326	244	164
electrical efficiency, %	38	45	55
overall efficiency (el. + thermal), %	90	85	90
Maintenance, $\text{€}/\text{kWh}$	1.5	2.5	0.5

According to the aforementioned, it is possible for the cost of the PEM fuel cell stacks to be lower than even 70  $\text{€}/\text{kW}_e$  in the near future, while a projected cost of only 40  $\text{€}/\text{kW}_e$  might be possible, assuming a power density increase to 4  $\text{kW}_e/\text{m}^2$  and the use of cheaper electrodes and bipolar plates. However, it is estimated that reducing costs to that level cannot be achieved with gradual improvements of the existing technologies, and besides new membranes, electrodes and bipolar plates production technologies and materials, even higher current densities and fuel cell efficiencies and lifetimes are required [2]. Furthermore, there is a trade-off between higher power densities and higher efficiencies, and depending on

capital costs and fuel costs there exists an optimal power density to minimize the cost per unit of energy produced. Current cells achieve  $0.3 - 0.6 \text{ A/cm}^2$  at  $0.6 - 0.7 \text{ V}$ , with power density in the range of  $1.8 - 4.2 \text{ kW}_e/\text{m}^2$  ( $2 \text{ kW}_e/\text{m}^2$ , average). Nevertheless,  $3 \text{ kW}_e/\text{m}^2$  are achievable with minor improvements, while values of  $4 - 6 \text{ kW}_e/\text{m}^2$  would necessitate improved membrane materials.



**Figure 4** Projection of PEMFC manufacturing and components costs [43]

But the most important role in the cost reduction of fuel cells is expected to be played by the manufacturing learning effects, due to higher production volumes. A quantification of learning effects (based on the concept of learning curves for the dependence of unit costs on cumulative production), and for an average learning factor equal to 0.8, predicts reduction of fuel cell system costs – for market entry value of  $15,000 \text{ €/kW}_e$  at  $20 \text{ MW}_e$  cumulative production – even below  $2,000 \text{ €/kW}_e$  at a cumulative production level above  $10,000 \text{ MW}_e$ , after an initial steep cost reduction to  $3,000 \text{ €/kW}_e$  at cumulative production of  $3,000 \text{ MW}_e$  (at this point certain applications become economically attractive) [3]. The projection of PEM fuel cell manufacturing costs, according to a scenario based on the learning effects of the corresponding industry, is presented in Figure 4 [43]. Especially for SOFC systems fuelled by natural gas, assuming annual production of  $500,000$  units, the cost is expected to range from  $725$  to  $1400 \text{ €/kW}_e$ , depending on the system size [2]. Table 7 compares the characteristics of conventional and SOFC systems, for a required capacity of  $200 \text{ kW}_e$ , predicting SOFC investment costs coming down to the level of conventional systems by 2030 [2].

#### Allowable Cost of Fuel Cell Systems

The cost at which fuel cells will become competitive to conventional systems, is determined by the corresponding costs of the competing technologies. Due to their higher efficiencies, fuel cell can withstand  $20 - 30 \%$  higher capital costs than other distributed systems, and this difference increases for smaller systems. Especially for domestic applications (up to  $5 \text{ kW}$ ), the high prices of household electricity is estimated to create an allowable costs of fuel cells up to  $2,000 \text{ €/kW}_e$  [3].

**Table 8** Performance and costs of PEM, SOFC, PAFC and AFC, up to  $5 \text{ kW}_e$  fuel cells [35]

	PEMFC	SOFC	PAFC	AFC
operating voltage, V	0.59 – 0.73	0.63 – 0.75	0.64 – 0.72	0.64 – 0.82
operating current density, $\text{A/cm}^2$	0.40 – 0.90	0.32 – 0.67	0.16 – 0.31	0.09 – 0.24
power density, $\text{W/cm}^2$	0.27 – 0.56	0.22 – 0.46	0.11 – 0.21	0.06 – 0.18
stack efficiency, % HHV	36.5 – 50.0	42.0 – 64.5	40.5 – 54.5	42.5 – 49.5
system efficiency, % HHV	23.0 – 31.5	27.0 – 41.5	26.0 – 35.0	27.0 – 32.0
total efficiency, % HHV	63.5 – 81.5	67.0 – 71.0	74.0 – 87.0	~ 87.0
lifetime, kh	7 – 21	15 – 59	30 – 53	4 – 8
, years	0.7 – 2.4	1.7 – 6.7	3.5 – 6.1	0.5 – 0.9

degradation, mV/year	13.1 – 74.5	28.0 – 73.6	14.9 – 39.4	78.8 – 254.0
, %/year	2 – 11	4 – 10	2 – 6	11 – 35
stack cost, €/kW <sub>e</sub>	300 – 900	200 – 600		150 – 600
system cost, €/kW <sub>e</sub>	530 – 1130	680 – 1080	2500 – 5000	375 – 825

The current technological status of small (0.5 – 5 kW<sub>e</sub>) PEM, SOFC, AFC and PAFC fuel cell stationary units, was recently reviewed by I. Staffell (Table 8). The system efficiency refers to electrical losses of the fuel cell system (fans, pumps, control) and the current conditioning unit (transformer, inverter), while total efficiency refers to both electrical power and heat cogeneration. Cost estimations, at the last lines of Table 8, refer to mass produced fuel cell systems according to today's state of the art manufacture technologies and materials, while current retail prices of demonstration fuel cell systems are referred in the range of €10,000 – 100,000 €/kW<sub>e</sub> [35].

**Table 9** Comparison of up to 5 kW fuel cell economics [35]

	PEMFC	SOFC	PAFC	AFC
estimated large scale production system cost, €/kW	300 – 900	300 – 600	2500 – 5000	325 – 675
target retail price, €/kW	220 – 420	510 – 970	660 – 1100	120 – 230
estimated lifetime, years	0.8 – 2.2	1.7 – 5.4	3.5 – 6.0	0.5 – 1.1
payback period, years	1.1 – 4.1	1.0 – 2.5		1.2 – 3.2

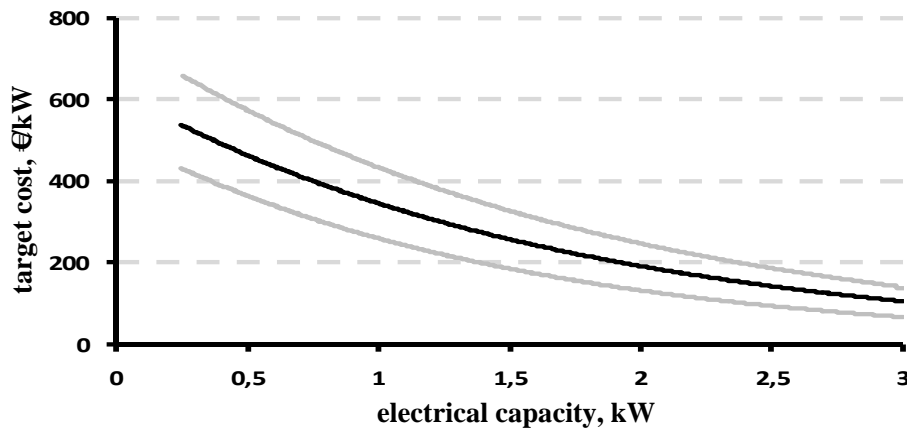
The utilization and the on/off cycles of a CHP system affect its potential benefit. Furthermore, higher overall efficiency leads to lower fuel consumption, while longer lifetimes lead to lower annualized capital costs. Concerning the relative influence of the demand profile, the nominal electric capacity, the efficiency and the lifetime of AFC, PAFC, PEMFC and SOFC systems, target costs were found to lie in a rather spread range. This spread was found to be crucially affected by the pattern of demand, and the amount of overlap between heat and power demand. Despite that fuel cell type did not substantially alter the annual savings, the difference in system lifetimes determined their target costs. The comparison between the estimated large scale manufacturing and target costs are summarized in Table 9.



**Figure 5** Estimated effect of fuel cell lifetime on the target cost for competitive commercialization [35]

Comparing the large scale production cost estimates with target costs for economic competitiveness, an indication of the market perspective of each fuel cell type can be figured out. Thus, according to the assumptions of Kendall et al, PAFCs and AFCs, under all circumstances, are expected to cost more than it is required to be competitive, while PEMs and SOFCs, exhibit an overlap between the estimated manufacturing cost and the target cost. Additionally, the estimated lifetimes of AFC and PEMFC were found shorter than their payback periods. PEMs have been surrounded by much commercial hype, and were responsible for much of the dramatic rise in interest in fuel cells over the last decade. Therefore, the majority of research and commercial activity worldwide is now focussed on PEMFC technology [52], giving the greatest potential to realize the improvements required to gain widespread usage. Estimates for stationary systems are more conservative due to the different design criteria, particularly the need for more

ancillary equipment, and a less aggressive power density. Many of the advantages of PEMFC are particularly suited to transport applications: e.g. the high power density, mechanical robustness and low operating temperature. However, application in the decentralized CHP market generates similar interest among manufacturers, as none of the above features is considered a disadvantage for stationary use, and overall performance appears to be similar to other low temperature fuel cells [35].



**Figure 6** Estimated effect of fuel cell capacity on the target cost for competitive commercialization [35]

System's lifetime (Figure 5) and the power demand (or the nominal electrical power output, of the required cell – Figure 6) were found to have the greatest impact on target costs, while the first is the main responsible for the differences in the target costs between the examined types. Kendall's results points out the considerable uncertainty in the cost targets for fuel cell CHP within a range of 300 – 700 €/kW, which seems quite typical [53]. Variations in electrical and total efficiencies among the fuel cell types, were found of little significance. In conclusion, mostly PEFC and SOFC have the potential to meet the allowable cost targets. It is critical however, the current technologies to obtain adequate production volumes [35]. High-temperature membranes, today, are expected to last not more than 20,000 hours and exhibit an overall installed fuel cell system cost of less than \$1,500/kW for initial commercialization (by 2008) and ultimately \$400/kW for large markets (by 2010) [46].

## 4.2 Fuel Cell Market Development

Fuel cell manufacturing costs and the directly related retail prices are expected to be strongly affected by the established production volumes and the corresponding learning effects. These volumes depend upon the rates of fuel cells commercialization, which, in turn, are expected to be determined by their price competitiveness. In this context, the fuel cell related industry, the most crucial leveraging factor for fuel the cell market development, is not only of major importance, but also represents an indicator of the situation so far and its near term future potential.

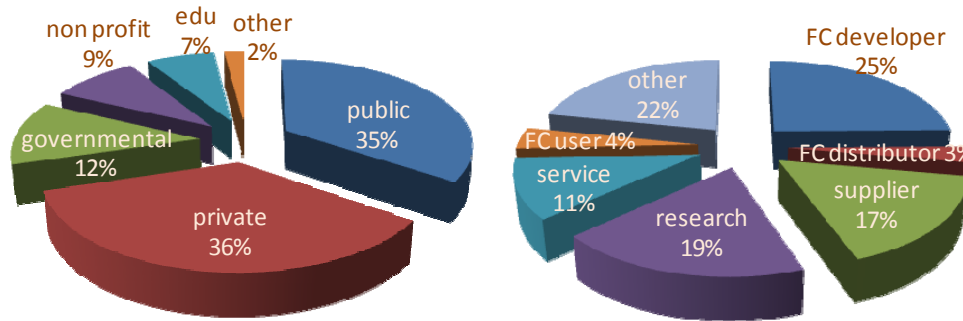
### Fuel Cell Industry profile

The commercialization of a new technology is a challenging and uncertain process. Likewise, the emerging fuel cell industry experiences numerous technical and market uncertainties to shift from primary Research and Development activities to production, marketing and sales. The pre-commercialization phase of a new technology is challenging and uncertain. Among others, the corresponding industry is challenged to manage long payback periods and a diversity of investment options. Today, the fuel cell market development process is quite possible to be passing through an initial fluid phase, characterized by a high degree of uncertainty concerning fundamental questions, like which technical alternatives will finally be accepted by the markets, or in how many years from now [54].

The 2006 Worldwide Fuel Cell Industry Survey [38] provides a profile of 181 organizations involved in the fuel cell industry, and reports on a variety of key performance indicators, the most important among which, in case of fuel cells, seem to be: sales and research & development expenditure. According to this survey, more than one-third of the aforementioned sample is private companies, while another one-third refers to public companies. Fuel cell developers and manufacturers correspond to a quarter of this sample,

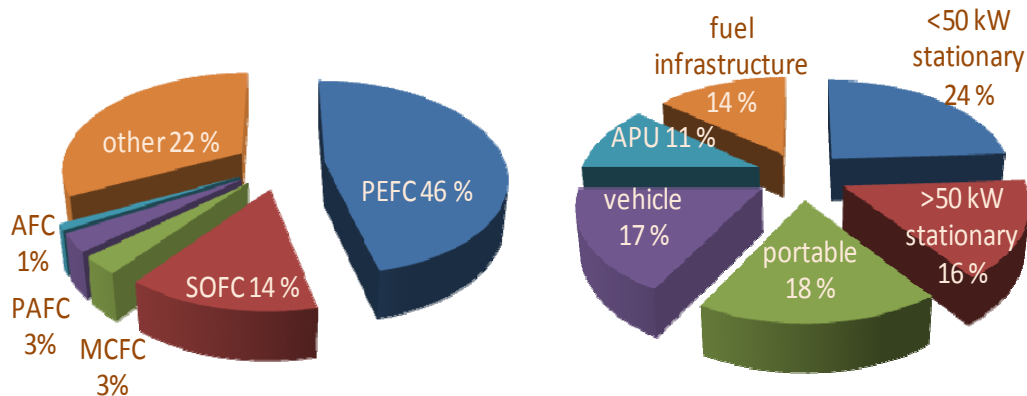


while about one fifth refers to research organizations and another fifth to suppliers towards the developers or manufacturers (Figure 7).



**Figure 7** Organization type and area of expertise of worldwide fuel cell related industry [38]

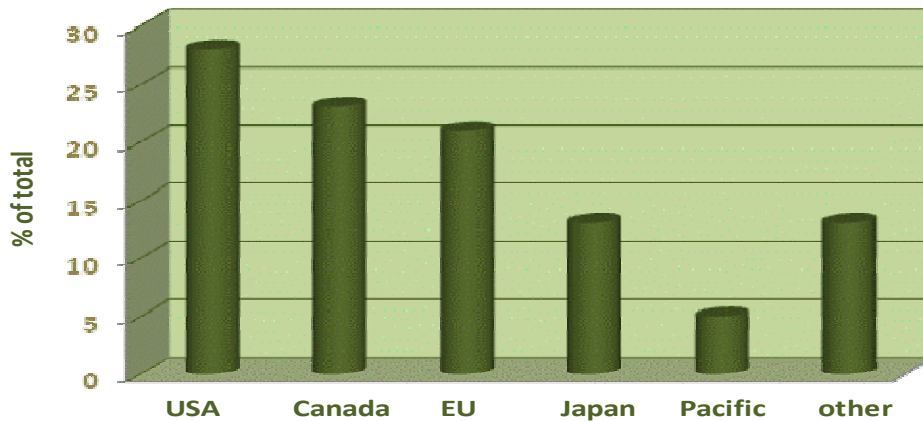
Fuel cell industry activities focus primarily on PEM fuel cells, followed by Solid Oxide Fuel Cells, Molten Carbonate and Phosphoric Acid Fuel Cell – Figure 8. Small (< 50 kW<sub>e</sub>) and medium (> 50 kW<sub>e</sub>) stationary power generation, were revealed as the main focus of almost 40 % of the respondents to the survey, followed by portable and vehicle fuel cell applications [38]. Proton exchange membrane fuel cells are the technology of choice for the transportation sector and also represent 70 – 80 % of the current small-scale stationary fuel cell market. Solid oxide fuel cells represent 15 – 20 % of the stationary market, and their share is projected to increase. Phosphoric acid fuel cells dominated the large-scale stationary market until 2002, but molten carbonate fuel cells are expected to take most of this market in the period 2005-2015, along with a gradually increasing share of SOFCs [2].



**Figure 8** Technology and market focus of worldwide fuel cell related industry [38]

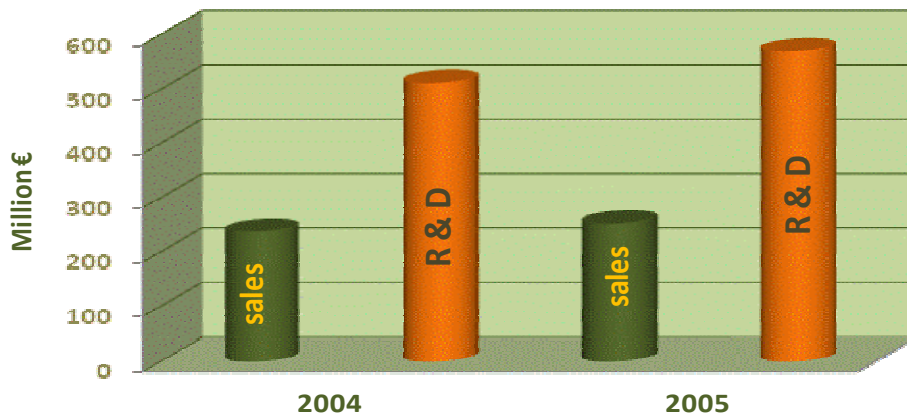
Almost 70 % of the respondents reported headquarters of fuel cell activities in United States and Canada, with the rest 30 % is shared by EU, Japan and Australia – Figure 9. Reported fuel cell related sales increased 7% (from 236 million € in 2004 to 252 million € in 2005) – Figure 10. Total R&D expenditure increased by 11%, from 2004 to 2005 (511 million € and 569 million €, respectively) – over 70 % allocated in North America (Figure 11), while the overall results suggest a growing industry, increasingly committed to the research and development needed for wide-spread fuel cell commercialization.

PricewaterhouseCoopers similar survey (referring to a sample of 23 fuel cell related industries), reports an almost 20 % increase of gross revenue (from 158 million € in 2004 to 190 million € in 2005), which corresponds to a restriction of the net losses of the fuel cell related activities by almost 20 % (from 321 million € in 2004 to 261 million € in 2005). It is worth to notice that the R&D expenditures are reported at 150 million €/a, and that the total assets of this industry sample were increased, from 2004 to 2005, by another 20 % (from 1.1 to 1.3 billion €) [37].



**Figure 9** Distribution of manufacturing and R&D activities [38]

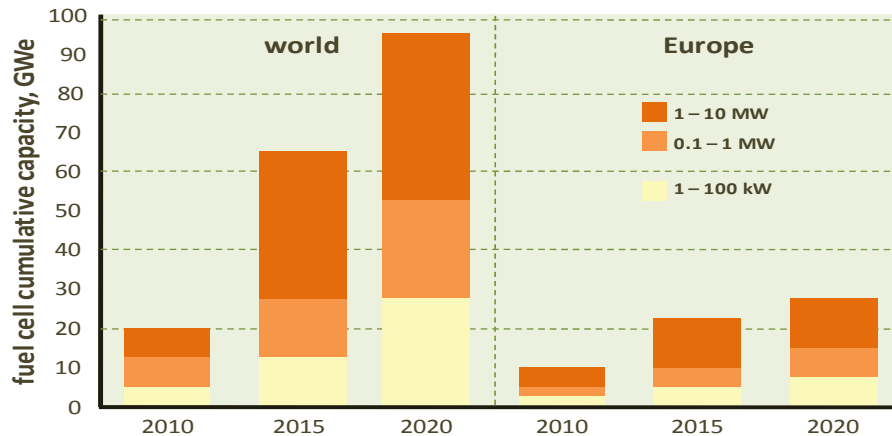
Most of the fuel cell related companies (or divisions of companies) are considered to be at development stage, focusing on research and testing. Although that might generate revenues from research contracts and services, few of them actually have market-ready fuel cell products. Those which report sales, they mostly refer to governmental demonstration projects and to a minor extend to military or other early adopters. Several of them, however, are already in the process of developing marketing and distribution networks, creating supply chain relationships and building up manufacturing capabilities for larger production volumes. Nevertheless, and despite the early stage of development strategic movements (mergers and acquisitions, alliances and collaborations, supply chain positioning etc) can already be detected, declaring a vividness and near future expectations [37].



**Figure 10** Comparative evolution of fuel cell industry sales and R&D expenditure [38]

Concerning the overall picture of the newly born sector, PricewaterhouseCoopers estimate that the general climate of uncertainty has been replaced by a keen focus to competitively priced products, fitting specific customer's needs, and which will have the potential to create returns for the industry. Furthermore, besides large corporate, many medium-small enterprises have shifted the heart of the industry towards the private sector [37,38]. Furthermore, FC firms face dilemmas in decision-making, regarding resource acquisition, competence development, product development and niche market selection. A new market development phase is said to be near, as FC firms are developing prototypes and pre-commercial products at an increasing rate. However, widespread commercialization has been announced several times and subsequently postponed an equal number of times [54]. There is a dynamic relationship among FC innovation, the market and the FC firms that emerge and compete on the basis of this new technology. Historical cases have shown that the development of a new technology brings about the development of a new industry in which entrepreneurs seize opportunities to innovate [54].

The FC firms are in a developmental phase and extensive R&D is conducted on the prospective technology. Nevertheless, the Hydrogenics 2004 annual report states that, for the fuel cell industry is still not clear when or whether a successfully completed R&D phase will lead to a commercially viable fuel cell power product. Nevertheless, FC firms are in the process of resource acquisition and proprietary competence formation, and facing severe challenges in R&D activities, that promises revenue generation through marketing and sales activities. To commercialize the technology, FC firms are challenged to shift from technology development to product development.



**Figure 11** UNEP 2002 forecast for fuel cell distributed power generation capacity [59]

At this stage, extensive investments are required for R&D and the gradual development of manufacturing capability. However, the market for FC products is almost non-existent as yet. PriceWatershouseCooper survey, reports that none of the corresponding companies were profitable in 2005, and that the financial performance reflects the costs of implementing strategies to refine pre-commercial technology, to develop product capacity and to secure market access [55]. Thus, the FC industry still appears not to be self-supporting and considering the long-term process of implementation. Return on investment is expected to take time. FC firms are facing long payback periods, and depend on subsidised projects, new venture funding or OEMs for capital to invest in further R&D. However, the FC industry exhibits technical progress and sales in early markets, suggesting potential in a seemingly unhealthy industry [54-56]. The successful commercialization of fuel cell is expected to require a substantial amount of investment and in the case of a start up company this often means many rounds of fund raising, with each succeeding round requiring more cash than the last as the company develops its product and expands its workforce. The financing of fuel cell ventures are expected to remain difficult for the foreseeable future [57].

### Fuel Cell Market Projections [3]

In 2002 it was estimated that above 1000 small (< 10 kWel) and almost 600 medium (> 10 kWel) fuel cell systems were in operation [58]. Today, fuel cell market surveys predict an increase in decentralized energy supply (regardless the technology), with fuel cells being one of the options. A summarization of their conclusion is presented flowingly [3]:

- on global scale, 95 GW of cumulative fuel cell capacity by 2020 (more than 50 % of the total distributed generation capacity) - Figure 5 [59]
- 150 GW installed capacity worldwide in the year 2020 [60]
- from 20 GW now to 280 – 350 GW in 2011, for decentralized electricity supply, worldwide, with progressing fuel cell contribution [61]
- distributed power up to 30 % of German electricity demand (one third by fuel cells) by 2015 i.e.
  - a potential of 400,000 heating systems/a – 20 to 70 % fuel cells corresponding to 1 to 5 GWeI
  - for commercial applications 12 GWe (10 – 50 % possible fuel cell share)

- for industrial applications 30 GWe (10-50 % possible fuel cell share) corresponding to a total fuel cell potential of 3 to 15 GW<sub>e</sub>, in Germany [62]
- fuel cells up to 3 % of German electricity demand by 2010 and 3.0 – 7.8 GW<sub>e</sub> by 2025[63]
- European MicroCHP up to 3.5 GW in 2010, corresponding to an increase in sales volume from 20 M€ in 2000 to 2 G€ in 2010 – (500,000 systems – 62 % share of fuel cells) [64]
- European MicroCHP total sales up to or 3.2 G€ (400.000 systems/a) in 2011[65]
- 110,000 domestic systems (mainly SOFC) by 2010 [66]
- 55,000 to 180,000 systems by 2010, and 1.75 to 5 million systems by 2025, in Germany (corresponding to 500.000 systems/a) [67]
- 230'000/a heating systems for new houses and 130'000/a for apartments by 2010 [68,69]
- 100'000 fuel cell systems in Germany and 250'000 in Europe in 2010 [70]

The aforementioned brief conclusions, reveal the uncertainty with respect to size and time scale of market introduction. Most of these market forecasts, however, coincide to a positive market sentiment. Concluding, for fuel cells there is still considerable uncertainty with respect to size and time scale of the market entry (quite possibly too late to make a contribution to Kyoto commitments for 2012), that may contribute to reluctance and postponing of investment. Stationary fuel cells may possibly shift the power range of electricity production to smaller capacities, as they achieve electrical efficiencies similar to combined cycle plants at much smaller size, and this might open new markets. As fuel cells have to succeed entering a functioning and fully developed market, cost is seen as the major market entry barrier. Today, stationary fuel cells are 2.5 to 20 times too expensive, with the balance of plant being responsible for a large share of total capital cost. Allowable capital costs in stationary applications vary between 800 €/kW<sub>e</sub> and – above 2000 €/kW<sub>e</sub> in some niche applications – with future electricity costs and the share of own consumption in total electricity production, being important parameters. The timing of fuel cell market entry, beside the cost, is determined by technical challenges that include reaching performance targets, increasing longevity, enhancing reliability of balance of plant, and adapting balance of plant components, e.g. gas reformers and micro turbines, to fuel cell systems [3].

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