1. Fuel Cells

AD Hawkes, L Exarchakos, D Hart, MA Leach
Centre for Energy Policy and Technology
Imperial College London, London SW7 2AZ
UK
a.hawkes@imperial.ac.uk

Ir. D. Haeseldonckx, Ir. L. Cosijns, Prof. Dr. Ir.
W. D'haeseleer
Department of Mechanical Engineering
KU Leuven
Belgium

1.1 Introduction

The fuel cell concept was first conceived in1839 by Sir William Robert Grove, who realized that reversing the process of electrolysis (splitting water to form hydrogen and oxygen using electricity) could result in a process that would create electricity. With the exception of some early experiments by the likes of Francis Bacon and Harry Karl Ihrig, Grove's discovery was largely ignored in favour of fossil fuelled engines until the National Aeronautics and Space Administration (NASA) pushed the technology forward in the 1960's. NASA adopted the technology over other contenders because of its relatively low weight and low toxicity. Present day space shuttles rely on fuel cells for electricity and water whilst in orbit.

Although there are several different types of fuel cells, they all operate on the same basic concept of electrochemical reaction of fuel and oxygen to produce water, direct current electricity, and heat. Fuel cells (essentially) consist of an anode, cathode, electrolyte, external electrical circuit, and fuel/air supply. Fuel is delivered to the anode, and an oxygen-rich mixture is delivered to the cathode. Ions migrate through the electrolyte, and electrons flow through the external circuit, creating the electrical current. A simplified cell is displayed in Figure 1. Fuel cell "stacks", which are a collection of a number of "cells", are incorporated into fuel cell "systems" which broadly consist of one or more stacks, fuel reformers, fuel and air pre-heaters, afterburner, recycling route, control system, power electronics including DC-AC inverter, etc.

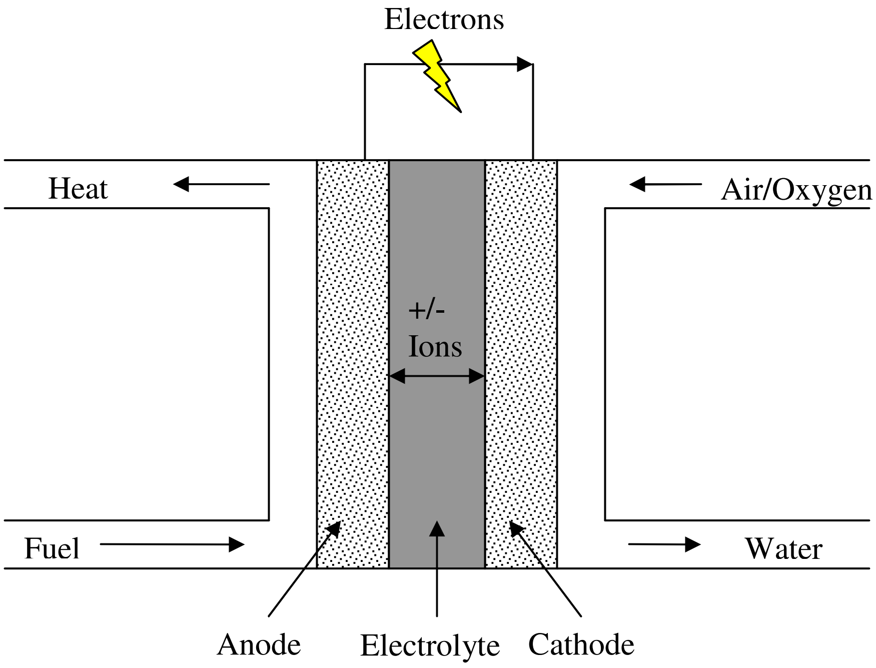

Figure 1. Simplified Generic Fuel Cell

The fuel cell differs from conventional heat engine technology (such as the internal combustion engine or the gas turbine), in that it does not rely on raising the temperature of a working fluid such as air in a combustion process. The maximum efficiency of a heat engine is subject to the Carnot efficiency limitation, which defines the maximum efficiency that any heat engine can have if its temperature extremes are known [1].

$$\text{Carnot efficiency} = \frac{T_H - T_L}{T_H},$$

where T_H is the absolute high temperature and T_L is the absolute low temperature. In contrast, the theoretical efficiency of a fuel cell is related to the ratio of two thermodynamic properties, namely the chemical energy or Gibbs Free Energy (ΔG^0) and the total heat energy or Enthalpy (ΔH^0) of the fuel [1].

$$\text{Fuel cell efficiency} = \frac{\Delta G^O}{\Delta H^O}$$

As the Gibbs Free Energy falls with increasing temperature, while the Enthalpy remains largely unchanged, the theoretical efficiency of the fuel cell falls with increasing temperature. Indeed, at high temperatures, the theoretical efficiency of a heat engine is higher than that of a hydrogen-fuelled fuel cell. However, because of the need for motion in a heat engine, either rotary or linear, there are significant materials issues associated with operating them at high temperatures, from the perspective of both durability and cost. Fuel cells do not have moving parts operating at high temperatures and thus are less susceptible to this problem [1].

In addition, other factors play a role in determining the actual efficiency of an operating fuel cell. For example, losses associated with the kinetics of the fuel cell reactions fall with increasing temperature, and it is often possible to use a wider range of fuels at higher temperatures. Equally, if a fuel cell is to be combined with a heat engine, for example in a fuel cell/gas turbine combined cycle, then high fuel cell operating temperatures are required to maximise system efficiency. All these factors mean that there is considerable interest in both low temperature and high temperature fuel cells, depending upon the application.

Some examples of commercial fuel cell projects can be found, but it would be premature to say that a market exists for the technology. Several demonstration projects are evident, and enormous research and development resources are directed towards improving the technology in order to push it into the market. As would be expected in any technology's R&D phase, most fuel cell projects are relatively small, ranging from 1kW$_e$ or less up to 250kW$_e$. There is no indication that these represent size limitations for fuel cells, which benefit from modularity.

Fuel cell types are classified according to the type of electrolyte utilised, geometry of construction, and temperature of operation (which relates back to electrolyte type). This report considers the five primary types of fuel cells; Solid Oxide (SOFC), Proton Exchange Membrane (PEM), Alkaline (AFC), Phosphoric Acid (PAFC), and Molten

Carbonate (MCFC). Direct Methanol Fuel Cells (DMFC) are considered to be a variant of PEM technology. Most of these technologies are considered to be pre-commercial, and therefore reported characteristics often represent a range of possible values from what has been achieved in practice so far, through to what is theoretically possible. Analysis is further complicated by the fact that no single source exists for comparison of life cycles of fuel cell types, and separate analyses define different boundaries to their LCA assessments. The combination of these factors results on reported LCA characteristics displaying wide ranges, and in some cases no information is available.

1.2 General Issues on Fuel Cell Technologies

Fuel cells are a cutting-edge technology that appears set to enter the broader energy market. They have the potential to achieve high electrical and overall efficiencies, sometimes with a wide range of choices of fuel. They are inherently modular, being constructed of individual cells connected together to form stacks, which can in turn be linked to create systems of virtually any size. They can be used to power small devices such as mobile phones, through a variety of CHP applications, and up to MW-scale power stations. Table 1 displays various proven or developing applications of reviewed technologies.

Table 1. Applications of various fuel cell technologies [1]

Fuel Cell Type	SOFC	PEMFC	DMFC	AFC	PAFC	MCFC
Domestic Power	☑	☑	-	☑	-	-
CHP	☑	☑	☑	☑	☑	☑
Large Scale Power	☑	-	-	-	-	☑
Transport	-	☑	☑	☑	-	-
Battery Replacement	☑	☑	☑	-	-	-

Fuel cell systems can also be categorised according to temperature of operation. Primary differences between operating temperatures are displayed in Table 2.

Table 2. Characteristics of High and Low Temperature Fuel Cells [1]

High Temperature Fuel Cells	Low Temperature Fuel Cells
Lower cost electrolytes	High cost precious metal electrolytes are common, although options are developing
Thermally constrained – slow start up, and slow dynamic response	Faster start-up times, and quick dynamic response
Few commercially available, with very high costs	Some commercially available, though costs are high
Fuel flexible – hydrogen and	Generally require pure hydrogen or

hydrocarbons	methanol
High operating temperature implies more useful heat is produced	Low operating temperature reduces value of waste heat
High temperature allows coupling with steam turbine, resulting in unprecedented electrical efficiencies	-
Resilience is a concern due to high temperatures	-
High temperatures implies exotic balance of plant materials are required, although intermediate temperature systems may circumvent this issue	Cheaper balance of plant

1.2.1 Peculiarities

The primary peculiarities of fuel cells in general are the presence of unusual materials, high part-load efficiency, modularity, lack of moving parts, high capital cost, and some degradation issues. They are also arguably unusual in the fact that they are relatively undeveloped, and therefore reported characteristics need to be considered with a wide range of uncertainty.

Materials

As noted below, exotic materials such a platinum, ruthenium, various ceramics, and alloys can all be found in various fuel cells. These materials are generally expensive, difficult and/or energy intensive to manufacture, and some are rare. A large portion of technical fuel cell research is directed at finding less expensive electrolyte, electrode and catalyst materials, but the challenge remains to demonstrably reduce costs to a commercially acceptable level.

Degradation

Degradation of fuel cell stacks is a significant hurdle to be circumvented or reduced before significant commercialisation of the technology is feasible. Degradation refers to the process by which voltage output (and thus power output) from the stack reduces in relation to the length of time it has been operating. The problem is apparent in all fuel cells, and particularly in those operating at high temperatures. Degradation rates as large as 5% per 1000 hours have been observed, and current best-practice is of the order of 1% per 1000 hours [2]. These degradation rates need to be overcome, particularly for stationary applications, where a minimum lifetime of 40,000 hours is expected. Load cycling and thermal cycling have been shown to accelerate the process of degradation, and sometimes lead to immediate performance reduction.

Part-load Efficiency

High part load efficiency is a beneficial aspect of fuel cell operation, allowing efficient operation at off design-point loads. Conventional thermal engines (e.g. IC engines and gas turbines) usually exhibit highest efficiencies at their design point near maximum load, but fuel cells can achieve high efficiencies across a wider range operating points.

Fuel cell efficiency curves are relatively flat across their operating range. This improves their potential for operation in application with lower load factors.

Modularity

Fuel cells benefit from intrinsic modular construction. Modularity implies that in principle, fuel cell systems with any power output can be produced by changing the number of cells per stack and/or changing the number of stacks.

Lack of Moving Parts and Reliability

Fuel cell stacks do not contain any moving parts. Therefore, they can operate relatively noiselessly and they are not liable to mechanical wear in the same sense as reciprocating machines. This improves their reliability. Of course, the absence of moving parts does not concern the balance of plant, which includes items such as blowers and pumps.

Capital Cost

As fuel cells are a developing technology, cost per kW_e installed are very high. Although spectacular cost reductions have occurred, and it is expected that costs will reduce further through learning-by-doing and scale-up of production, commercially competitive technology is yet to emerge. Substantial research effort is being directed at reducing costs, including the DOE's Solid State Energy Conversion Alliance (SECA) which plans to bring stack cost down to US\$400/kW_e, although there is some doubt in the fuel cell community as to whether or not this target can be achieved in the short to medium term.

1.2.2 Environmental Aspects

Fuel cells are generally perceived to be an environmentally friendly technology. With the exception of the specialised materials mentioned above, fuel cells are manufactured from readily available materials, and manufacturing processes are not remarkably different from those of other thermal energy conversion devices.

Fuel cells of various types operate on a number of fuels, from PEMFCs and AFCs which are fuelled purely by hydrogen, through to SOFC and MCFC systems that are adaptable to virtually any hydrocarbon fuel. Pollutant emissions are virtually zero during operation of all fuel cell types, and carbon dioxide emissions vary between fuel types employed, but are always at least competitive with other modern conversion technologies utilising the same fuel. For example, while fuel cell cars powered on gasoline reformer technology will have little or no benefit in reduced greenhouse gas emissions, methanol-powered cells may produce 25% less CO_2. Using hydrogen produced from natural gas could result in cuts of up to 40% (in the automobile sector), while hydrogen from biomass, potentially a closed-cycle process, could have net CO_2 emissions of zero. In the ultimate renewable/zero-emission scenario, fuel cells can be run on pure hydrogen produced from renewable energy, using electrolysis of water [1].

Figures 2 and 3 display pollutant emissions and carbon dioxide (CO_2) emissions for various fuel cell technologies based on manufactures goals and prototype characteristics [3]. CO_2 emission rates reported [3] are for electricity-only operation (i.e. no credit for heat output in CHP operation). Figure 2 demonstrates the low pollutant emission characteristics of fuel cells. For example, NO_x emissions range from 0.014 to 0.027 kg per MWh electricity, whilst CCGT and coal-fired power stations produce approximately 0.3 and 3.5 kg/MWh respectively [4]. NO_x from power generation are predominantly formed due to fixation of atmospheric oxygen and nitrogen, which occurs at high temperatures such as those experienced during combustion. Low NO_x rates observed in fuel cells are related to their intrinsic avoidance of combustion. Even high temperature fuel cells are not hot enough to produce significant NO_x.

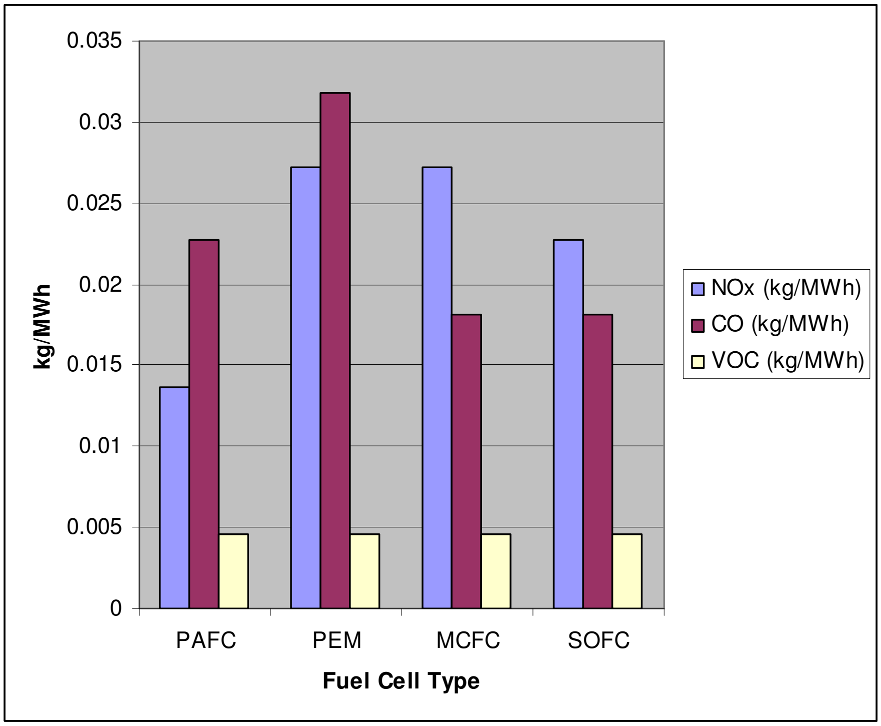

Figure 2. Selected Pollutant Emissions for Various Fuel Cell Types [3]

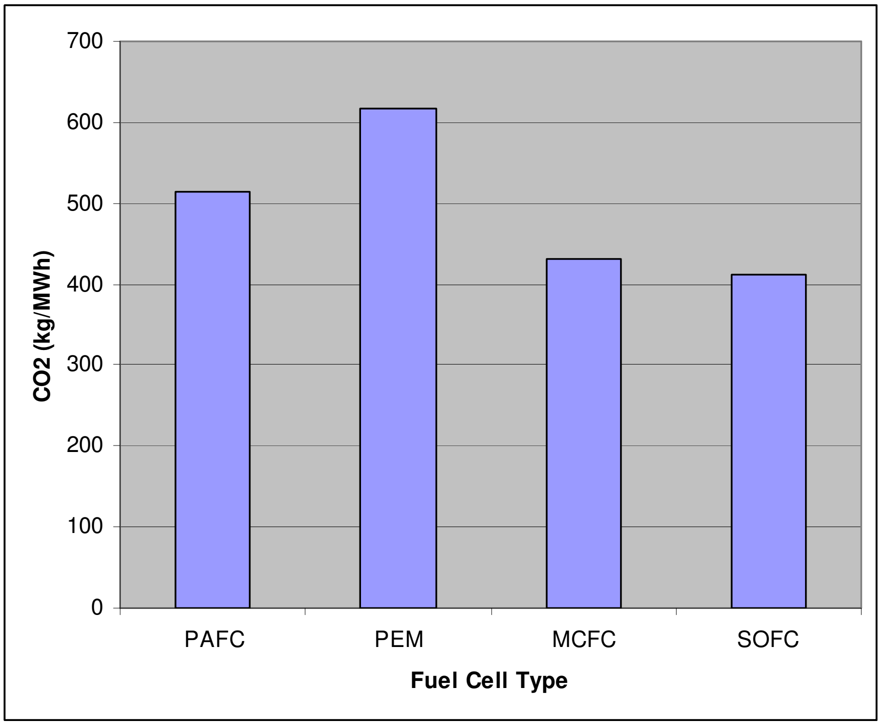

Figure 3. CO2 Emissions for Various Fuel Cell Types [3]

Note that carbon dioxide emission rates in Fig 3 are based on natural gas being utilized as the fuel source, with or without pre-reforming technology depending on the fuel cell type. CO_2 emissions are strongly dependent on the type of fuel employed.

1.3 Description of Fuel Cell technologies

1.3.1 Solid Oxide Fuel Cell

Solid Oxide Fuel Cells (SOFCs) are high temperature fuel cells that use a solid electrolyte to conduct oxygen ions. The two geometries of SOFCs that are being developed are (a) tubular and (b) planar types. The tubular configuration is exclusive for SOFCs while planar configuration is used in all other types of fuel cells. SOFCs under development may also be classified into types of electrolyte, and temperature of operation. Electrolyte types are discussed below, and the two general temperature ranges are intermediate temperature (approx 650 – 800^{o}C) and high temperature (approx 800-1000^{o}C). The cell is constructed with two porous electrodes that sandwich an electrolyte. Air flows along the cathode. When an oxygen molecule contacts the cathode/electrolyte interface, it acquires electrons from the cathode. The oxygen ions diffuse into the electrolyte material and migrate to the other side of the cell where they contact the anode. The oxygen ions encounter the fuel at the anode/electrolyte interface and react catalytically, giving off water, heat, and electrons. Carbon dioxide is another product of SOFC operation where hydrocarbon fuels are employed. The electrons transport through the external circuit, providing electrical energy [5]. The concentration of reactants at the anode and cathode influence the efficiency of the cells; referred to as concentration polarisation. Therefore, at part load, fuel cells are efficient, because reactants are not depleted on anode and cathode surfaces as rapidly as at full load.

SOFC electrolyte materials are ceramics; Yttrium Stabilised Zirconia (YSZ) is the most commonly used material for electrolytes and it offers high ion conductivity at temperatures exceeding 700^oC with minor electronic conductivity (but above 1500^oC it becomes an electron conductor, creating a short circuit in the cell). It is preferable to operate SOFCs at lower temperatures to improve structural durability and to avoid temperature limitations of affordable balance of plant components. The materials used in cell construction have different thermal expansion coefficients, which causes difficulty in preserving stack construction integrity and leads to start-up times approaching 24 hours in developing technology. Intermediate temperature SOFC systems are being developed to achieve low temperature operation, circumventing many of these problems. However, higher operating temperatures (700 to 1000^oC) offer improved internal reforming characteristics, expanding the possibilities for type of fuel utilised and creating a broad range of potential CHP applications. The flexibility of fuel use is reflected by the fact that CO and hydrocarbons (such as methane) can be used as fuels.

The effects of increasing pressure, as is the case for PAFCs and MCFCs, are positive for the cell performance. Temperature, fuel composition and fuel utilization all influence the performance of cells. As with all fuel cells to a varying degree, performance and lifetime are also influenced by impurities in the fuel supply. HCl, $NH_{3,}$ and H_2S can cause degradation or sudden performance drops, but levels lower than 1ppm, 5000ppm, and 0.1ppm respectively appear to eliminate detrimental effects [5].

Because SOFCs operate at high temperatures, and therefore lend themselves to applications in which this high-temperature thermal energy can be used. This heat can be used in two basic ways – for heating processes such as those in industry or for integration with turbines for additional electricity production. Until recently the concept of adding a turbine cycle to the fuel cell system would have necessitated a system size of well over 1MW. However, recent advances in microturbines have led to the concept of a combined power plant of 250kW with an electrical efficiency approaching 70% [1].

1.3.2 Polymer Electrolyte Membrane Fuel Cell

PEMFCs have high power density, rapid start-up and low temperature operation. This makes them ideal for use in transport, battery replacement, and load following operation (although load following still negatively impacts durability). The present cost of PEMFC systems is high, and does not assist its cause in the stationary power generation market. However, mass-manufacturing techniques are expected to bring down those costs and may enable PEMFC to compete with conventional generators and other fuel cells in the medium term. Domestic heat and power production is a useful example. Although PEMFCs operate at about 80°C, this is sufficient for space heating and hot water requirements. Several demonstration and some commercialisation projects are evident [1].

Polymer Electrolyte Membrane Fuel Cells (PEMFC's) operate in a temperature range of 60-80^{o}C and use a solid, proton-conducting fluorinated sulfonic acid or similar polymer as electrolyte, such as Nafion from DuPont. This solid electrolyte allows a compact construction of the fuel cell and also allows more straightforward operation under pressure, which can improve the performance considerably [5].

The catalyst commonly used in PEMFCs is made of platinum with carbon or graphite as structural supportive materials. The drawback of the use of platinum is the fact that CO concentration of even 10 ppm causes performance degradation of the cells. However, the combination of Platinum/Ruthenium in the electro-catalyst, improves the tolerance. Similarly to most fuel cells, sulfur must be kept at low levels in the fuel stream.

Since PEM-developers are mainly focusing on the residential, automotive, and tertiary market, natural gas is the most obvious fuel to use. This implies that the development of small-scale, affordable and high-performance reformers is inherently coupled to the development of PEMFCs [5].

1.3.3 Direct Methanol Fuel Cell

The Direct Methanol Fuel Cell (DMFC) is closely related to PEM systems: the structure of the fuel cell is almost identical and Nafion has often been used as a membrane. DMFCs overcome a perceived barrier for PEM-systems; the storage and production of sufficiently pure hydrogen presents many challenges. Therefore, the possibility to use methanol directly as a fuel has been studied. The advantages of methanol are a 6 times smaller storage capacity compared to hydrogen required for a same amount of energy, the possibility to use liquid storage at atmospheric pressure, and no need for the construction of hydrogen infrastructure. Methanol undergoes electrochemical reaction with water to CO_2 at an operating temperature of 80 to 130 °C. This low temperature makes short start-up time possible.

The main drawbacks of the DMFC are the excessive use of expensive materials like platinum (up to ten times more than high performance PEMFCs) and ruthenium, and the toxicity of CH_3OH. Low cell voltage, resulting in low current density, is caused by cross-over of neutral methanol from anode to cathode side.

Compared to other fuel cell types, the development of the DMFC has only just begun. Initial developments suggest that the DMFC will be used as a substitution for the classical batteries in small scale, portable appliances (e.g. cell phones, laptops, CD players, etc). In June 2004, Toshiba presented 'the smallest fuel cell in the world', which could feed a CD player for 20 hours. Another market which shows great interest in the DMFC is the automotive market. This product is still at the research and development stage, but has a very large estimated potential [5].

1.3.4 Phosphoric Acid Fuel Cell

Together with the SOFC and the Molten Carbonate Fuel Cell (MCFC), the Phosphoric Acid Fuel Cell (PAFC) is one of the high temperature fuel cells. Its operating temperature is approximately 200 °C and it uses concentrated phosphoric acid as the electrolyte. The oxidant for terrestrial applications is air and the cell usually operates at ambient pressure. The electrodes' most important material is platinum but the use of high surface area graphite structurally supporting the platinum has resulted to significant reduction of the platinum loading without reducing the electrodes' performance.

Although PAFC systems have been successfully integrated into buses, they are not ideal for transport applications. They have been relatively successful, in fuel cell terms, as stationary cogeneration plants, with many PC25 units installed and operating worldwide. The PC25-module by International Fuel Cells/ONSI (now UTC Power), has a power output of 200 kW_e and 220 kW_{th} and an electrical and thermal efficiency of 40% and 45% respectively. Since 1991, over 265 PAFC units have been installed by UTC Power [6]. Typical applications lie in hospitals and leisure centres, where the waste heat can be used in laundry/water heating, and in other areas where consistent and reliable power is required. The PAFC has historically been the most successful fuel cell type, but in recent years MCFC technology has been leading in terms of sales. Cost reduction below €3000/kW_e for PAFC technology appears to be difficult, and active manufacturers are focusing on extending the lifetime of the product.

The high operating temperature makes the PAFC suitable for some CHP-appliances, but also results in slow start-up times of 3 to 4 hours. The main disadvantages of a PAFC-system are the sensitivity of the anode to CO and sulphur-poisoning, the use of platinum as a catalyst and the use of a liquid electrolyte, which complicates the prospect of operation under pressure [5].

1.3.5 Molten Carbonate Fuel Cell

The Molten Carbonate Fuel Cell (MCFC) is a high temperature fuel cell which uses molten carbonate salts as its electrolyte. Its operating temperature is around 650 °C. This technology has attracted interest because the high temperature makes a separate fuel reformer redundant, and makes heat recuperation and steam production possible for industrial and other applications. Furthermore, compared with the PAFCs, its higher temperatures allow higher overall system efficiencies and increased flexibility of fuel choice. However, the high temperature can cause corrosion to system components and slow start up times, through similar mechanisms to that of the SOFC systems.

The MCFC is well developed technology, and is already commercially available. FuelCell Energy [7] and MTU [8] are amongst the foremost developers in the market. Subsequently, the MCFC is often referred to as 'the second generation' fuel cell (after PAFC). Prototypes in a power range of 250 kW_e to some MW_e are being operated successfully. The mentioned power ranges illustrate the suitability of the MCFC-technology for large industrial appliances. However, the start-up time, which can be up to 20 hours, could be improved [5].

MCFCs are likely to occupy the same market segment as SOFCs. They run slightly cooler at 650°C, making their combination with a gas turbine less straightforward. Whether or not high temperatures would bring about materials problems is an important issue. The primary difference between the MCFC and the SOFC is the need for CO_2 recirculation in the MCFC system, meaning that it is difficult to design one below about $250kW_e$ due to an overly complex manifolding arrangement. This makes market entry in residential CHP difficult [1].

1.3.6 Alkaline Fuel Cell

The Alkaline Fuel Cell (AFC) belongs, as the PEMFC and the Direct Methanol Fuel Cell (DMFC), to the class of low temperature fuel cells. Its operating temperature is approximately 70 °C and it uses liquified potassium hydroxide as an electrolyte. The big advantages of this system are its very short start-up time, less use of expensive materials and high efficiencies compared to the other fuel cell types. The main disadvantage of the AFC is probably its sensitivity of the potassium hydroxide to CO_2-poisoning and of the catalyst to CO-poisoning. This sensitivity implies that air-fuelled AFC systems must include CO_2 scrubbing, and the hydrogen fuel stream must be free on CO. Similarly to PEMFC systems, AFCs low operating temperature makes this fuel cell type less practical as a CHP-system.

The alkaline fuel cell, as mentioned, has a long history in space programs, because as it was the first to be adequately developed. It is still used in the space shuttle to produce power for the on-board systems by combining the pure hydrogen and oxygen stored in the rocket-fuelling system, and producing water for the astronauts to drink [1]. It has recently undergone something of a renaissance because it can be made cheaply in comparison with other currently available fuel cells, though mass-manufacture of other cells may reduce this advantage. Only a handful of companies are currently working in the field of AFCs [5].

1.3.7 Future Fuel Cell Technologies

1.4 Present Fuel Cell Market

In general, the current fuel cell market is limited to a few commercial products with high capital cost. MCFC technology is leading in terms of recent installations for large stationary applications, but it would be premature to suggest a competitive commercial market exists. Results of a PWC survey on worldwide fuel cell activities [9] reported total sales of \$240M in 2002 rising to \$338M in 2003. Approximately 11% of this activity was in Germany. Overall, activity in Europe is substantially less than that of the United States or Japan. Contribution by fuel cells to the generating mix is very small in Europe.

The total number of large stationary (>10kW) fuel cell installations worldwide was just under 800 in late 2005, with MCFC technology by FuelCell Energy achieving the most installations in 2005. However, although SOFC technology remains the least developed in terms of number of installations, it has the largest number of companies actively researching its improvement [10].

Fuel cell micro-CHP appears to be a market attracting much attention from developers, with the potential to result in significant aggregate penetration of the technology. PEM and SOFC technology in the 2-5kW$_e$ range are the primary contenders, although there is also some limited interest in AFC technology. European researchers are more active in the development of residential micro-CHP rather than large stationary applications, with about 20% of the world's operating units (the rest are divided almost equally between the USA and Japan, with the rest of the world also achieving a small share) [11]. Micro-CHP is generally regarded to be an important early market for fuel cells.

1.5 Future Development

As fuel cells are a developing technology just entering the first stages of commercialisation, it is difficult to draw broad conclusions regarding future scenarios. Current trends, which may be indicative of future outcomes, suggest that SOFC and MCFC technology will be important technologies in the large scale stationary energy market. The lower temperature, less fuel-flexible technologies (PEM, AFC, and possibly PAFC) may be more important at the small-scale end of the stationary market, particularly in residential applications. It appears the SOFC technology may also play a role at the residential level, although the withdrawal of Sulzer Hexis from this market is a set back.

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