

Fuel cells – a 21st century power system

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Abstract

Fuel cells have been applied for practical power generation in spacecraft since the first manned flight. Because of their wide range of potentially attractive characteristics and supported by the oil shocks of the 1970s as well as growing environmental concerns, their development for terrestrial applications has been pursued ever since. Today the first wide-spread demonstrations are taking place. This paper will give a review of the principles, characteristics, construction, system lay-outs and development status of fuel cells. The prospects for integration in energy supply infrastructures and further commercialisation will be discussed. © 1998 Elsevier Science S.A.

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1. Introduction

Fuel cells are related to batteries. Both convert chemical energy directly into electricity. The difference is that in a battery the chemical energy has to be stored beforehand, while fuel cells only operate when it is supplied from external sources. The fundamental mechanism of fuel cell operation is the inverse water hydrolysis reaction (see Fig. 1). Catalytic oxidation of hydrogen at an anode and reduction of oxygen at a cathode create a potential difference between those electrodes. This can be exploited in an external circuit if an insulating electrolyte between the electrodes allows for ionic mass- and charge transfer. The product is then water and the chemical energy of the reaction is liberated as electricity and, because of polarisations and ohmic losses, heat.

This process is not subjected to the Carnot principle and its related limitations. As a result, not only total efficiencies but also the electrical efficiency can be high. This is true over a wide range of installed capacities and part-load operation. In the latter case, the efficiency of the fuel cell proper even increases further with decreasing part-load to a relatively low value.

The characteristic efficiency properties allow for modular construction with clear advantages for optimising QA and QC both in the factory and on-site. Moreover, this provides direct cost savings as on-site construction times are minimised and capacity can be installed and added as necessary and installation of overcapacity is avoided.

Because the fundamental reactions can be catalysed by a

variety of materials at different temperatures, fuel cells have been developed for a number of operating temperatures ranging from room temperature to 1000°C. The selection of the type of fuel cell will, therefore, be determined by the intended application.

Added versatility derives from the flexible heat to power ratio which may be as low as 0.2–0.3, and the flexibility regarding primary fuels. In fact, any fuel will do from which a hydrogen-containing gas mixture can be produced. However, the conversion to hydrogen causes some efficiency loss. Clean-up of the fuel or product gas may be necessary. Air generally constitutes the other reactant. Due to the necessary gas clean-up for the fuel processor and (in general to a lower degree) also for the fuel cell proper, as well as to the fact that the electrochemical reactions in the cell are very clean, fuel cell plants show extremely low emission levels.

Added environmental benefits are due to the absence of moving parts in the fuel cell proper; noise and vibration levels are caused by auxiliary equipment and can be minimised. In addition, this makes fuel cells in principle highly reliable and low in maintenance cost.

High temperature fuel cells have some additional advantages: plant simplification and cost savings because H₂/CO mixtures can be used as fuel, fuel processing may take place in the cell itself, and the possibility for integration with steam or gas turbines. The latter will enable electrical efficiencies up to 70% LHV, although it further complicates the auxiliary equipment.

Thus, fuel cells have a wide range of attractive characteristics and potential applications.

2. Fuel cell types

Fuel cells are identified by probably their most important component, i.e. the electrolyte. The electrolyte determines the operating temperature and with that the catalysts to be applied in the electrodes, as well as the process gas requirements.

In the alkaline fuel cell (AFC) a concentrated solution of potassium hydroxide acts as the electrolyte as well as coolant. It conducts hydroxyl ions from cathode to anode, operates at about 80°C and is sensitive to CO₂ poisoning.

The polymer electrolyte fuel cell (PEFC, PEMFC, SPFC) also operates at low temperature (80–100°C). The electrolyte is a solid polymer, which, when hydrated, conducts protons from anode to cathode.

Both types of fuel cell operate at a temperature level that necessitates the use of platinum, generally dispersed on carbon black, as a catalyst in the electrodes. Platinum at these temperatures is highly sensitive to poisoning by CO.

The same is true for the phosphoric acid fuel cell (PAFC) which operates around 200°C. The electrolyte also conducts protons from anode to cathode and is kept in place by an inert porous structure. Both PAFC and PEFC are tolerant with respect to CO₂.

The molten carbonate fuel cell (MCFC) employs a molten mixture of alkali carbonates as an electrolyte which provides for mass- and charge transfer through carbonate ions from cathode to anode and is kept in place by an inert porous structure. CO₃²⁻ transport necessitates the addition of CO₂ to the cathode feed gas, which is then liberated again at the anode. The operating temperature is around 650°C which allows the use of catalysts like nickel in the electrodes.

The electrolyte in solid oxide fuel cells (SOFCs) is generally yttria-stabilised zirconia, a solid which conducts oxygen ions from cathode to anode at high temperatures: around 1000°C. The electrolyte as well as the electrodes are ceramic materials.

Fig. 2 summarises the most important features of the different fuel cell types.

Both the MCFC and SOFC operate at temperatures high enough to promote the water gas shift reaction at the anode, thus enabling H₂/CO mixtures to be used as fuel feed stock. Also certain fuels can be processed directly in their anode chamber, e.g. reforming of natural gas. In the case of MCFC this requires an additional catalyst. The advantages are a lower cooling need and a higher efficiency.

3. Construction

For ease of manufacturing of single components and of assembly of those components into a cell, fuel cells are generally constructed according to a flat plate design. An exception is a tubular design in case of the SOFC. Active electrode areas range up to 1 m² for PAFCs and MCFCs but are smaller for the other types. The combined thickness of electrodes and electrolyte is seldom more than a few millimetres.

The electrodes are electric conductors with a high porosity, the latter for maximising the electrocatalytic surface area and to enable transport of process gases to and from the catalytic sites. The reactions take place at the interface of catalyst and electrolyte, so sufficient contact area has to be provided in this respect. The electrolyte, apart from being an electrical insulator and ionic conductor, should also act as a gas barrier between the electrodes.

Operating fuel cells produce direct current with densities

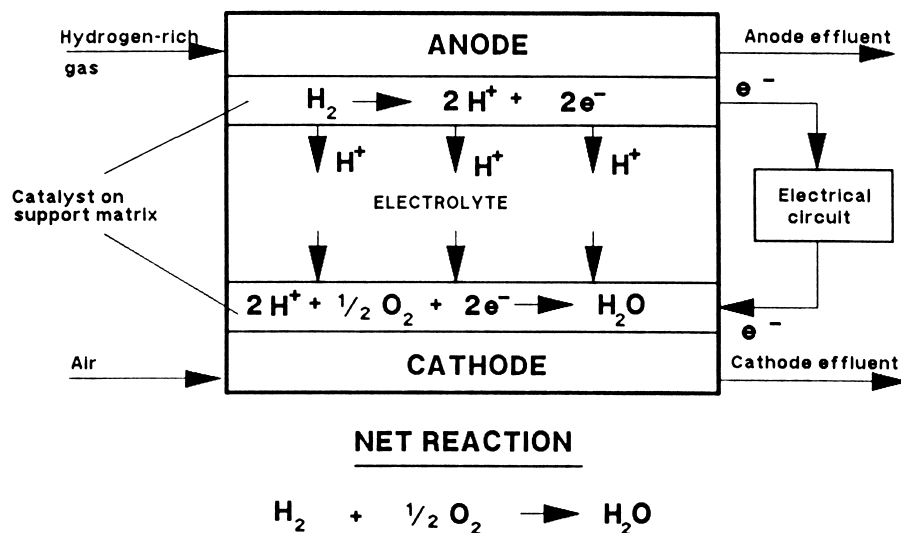


Fig. 1. The principle of fuel cell operation (PAFC).

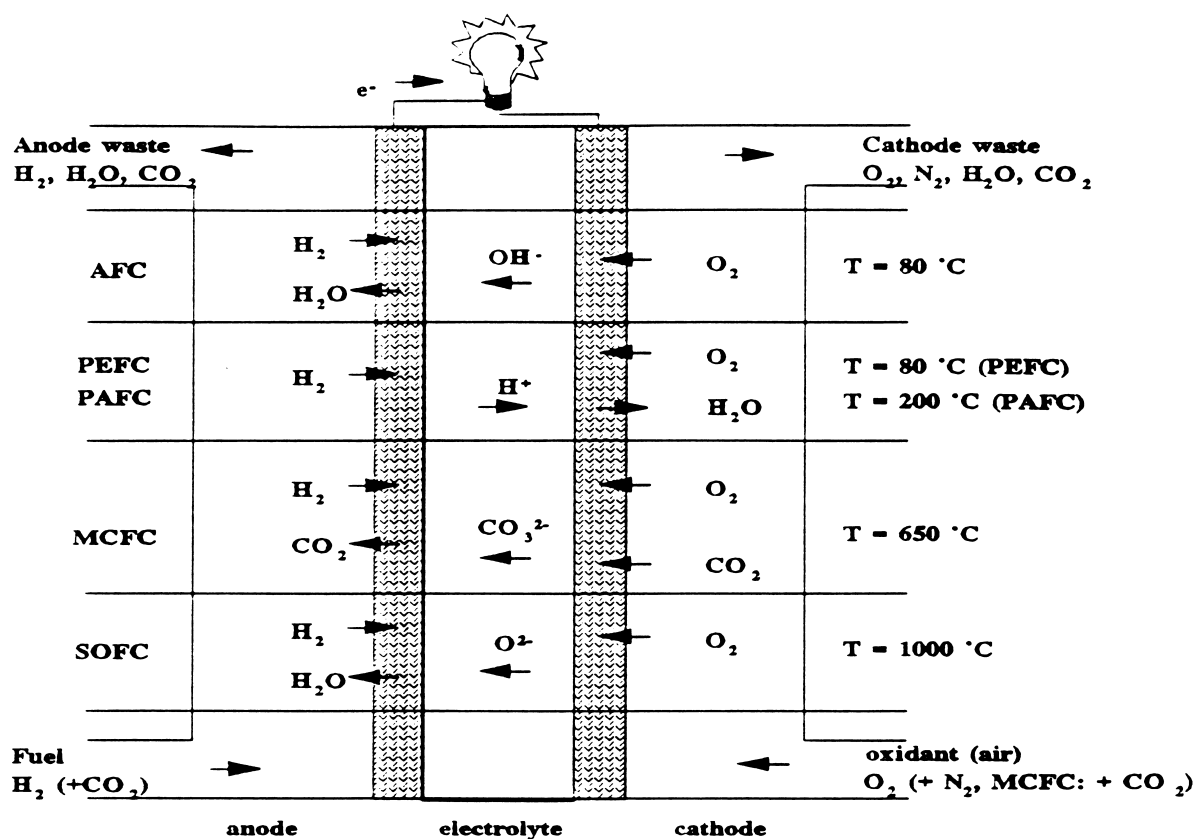


Fig. 2. Electrochemical reactions for different types of fuel cells.

that in extreme cases may reach several A/cm^2 . Single cell voltages, however, are in the range of 0.5–1 V. For practical purposes, therefore, they are connected in series.

In a planar design, series connection can easily be accomplished by stacking of individual cells with a bipolar plate in between. This plate provides the electrical contact between the anode of one cell and the cathode of the adjacent cell, while keeping their process gases separated. The plates are shaped to enable gas supply to, distribution over and removal from cells (see Fig. 3). Construction materials are metals, graphite or conducting ceramics.

Dependent upon stack design, any flow configuration can be realised, i.e. co-, counter- and cross flow in the anode and cathode compartments, respectively. They all have in common a lateral flow over the electrodes, resulting in depletion of active species and enrichment of reaction products towards the cell outlet. This, in turn, creates lateral current density, voltage and especially temperature gradients. The latter may be as high as 100°C in high temperature cells and have to be kept within limits by adequate cooling.

4. System configurations

From the above it will be clear that for practical applications fuel cells will need substantial peripheral equipment. Fuel cell systems consist, therefore, in general, of the fol-

lowing four sections (see Fig. 4): fuel processing, power generation in the fuel cell stacks, power conditioning and heat recovery and/or power generation in integrated gas and steam turbines (driven by the exhaust gases of the fuel cell and the fuel processing sections).

The provision of a hydrogen-rich fuel gas to the fuel cells usually requires the primary fuel to undergo pre-processing steps. Reforming of natural gas, naphtha or gasification of coal and biomass is, in general the first step to produce a hydrogen-rich gas with also high contents of carbon monoxide and carbon dioxide.

The low temperature fuel cell types (AFC, PEFC and PAFC) need, however, high purity hydrogen, whereas the high temperature fuel cells are capable of oxidising carbon monoxide to carbon dioxide inside the anode simultaneously with the electrochemical reactions. Therefore, low temperature fuel cell systems comprise a second pre-processing step called the 'water gas-shift reaction' in which carbon monoxide reacts with steam and is converted into hydrogen and carbon dioxide. These processes need relatively high temperature heat and must be integrated into the system. When these pre-processing steps are performed in separate reactors, the fuel cell system is referred to as an 'external reforming' system. This is always the case for the low temperature fuel cells.

The term 'internal reforming' describes those fuel cell systems where the reforming reactions take place in the

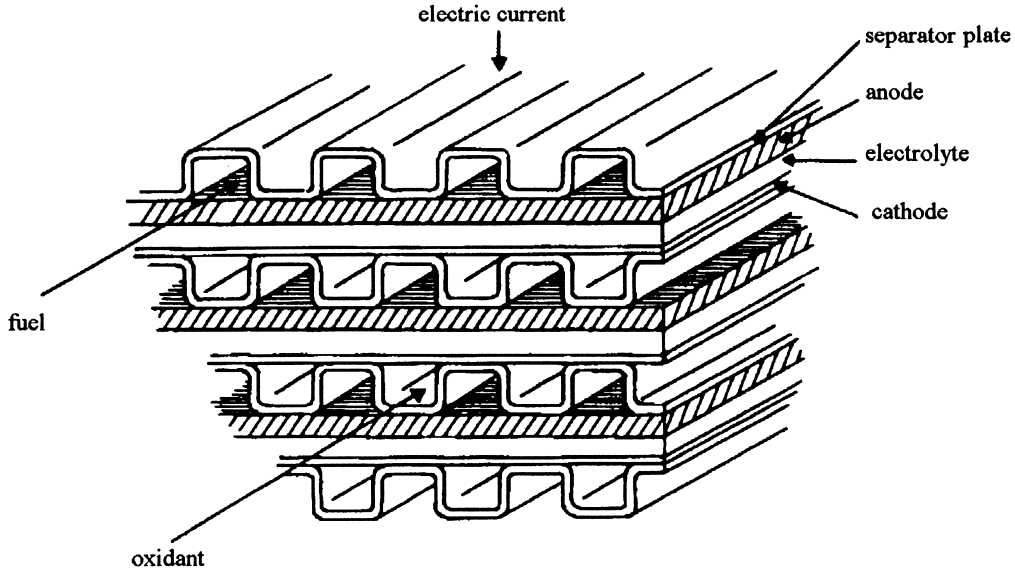


Fig. 3. Example of stacking single cells.

fuel cell anode almost simultaneously with the electrochemical reactions. The internal reforming concept is feasible in high temperature fuel cells because the steam reforming reaction can proceed (in the case of MCFCs with the help of a separate catalyst), due to the production of water via the anodic oxidation of hydrogen. The internal reforming concept eliminates the need for the external fuel processor with its ancillary rotating and heat exchanging equipment and, therefore, results in a highly efficient, simple, and cost-effective system even for small capacities.

The electrochemical power generation takes place at the level of individual cells, which produce direct current. The cells are stacked together to achieve a desired voltage which is then converted with a high efficiency to alternating cur-

rent in the power conditioning section. The primary fuel energy not utilised for electric power production is available as waste heat and needs to be removed from the fuel cell in order to maintain the isothermal condition. Heat removal from the stacks is generally accomplished by making use of the process gases, especially through high cathode flow. Through integration, this waste heat is used in other parts of the system, e.g. to produce the hydrogen from the primary fuel. Separate cooling circuits may be applied also for which in AFCs the electrolyte and in PAFCs a pressurised water cycle is used, whereas for high temperature fuel cells an additional option is to reduce the cooling needs by incorporating, e.g. a reforming step in the stack. The remaining part of the waste heat is used to produce steam for process heat

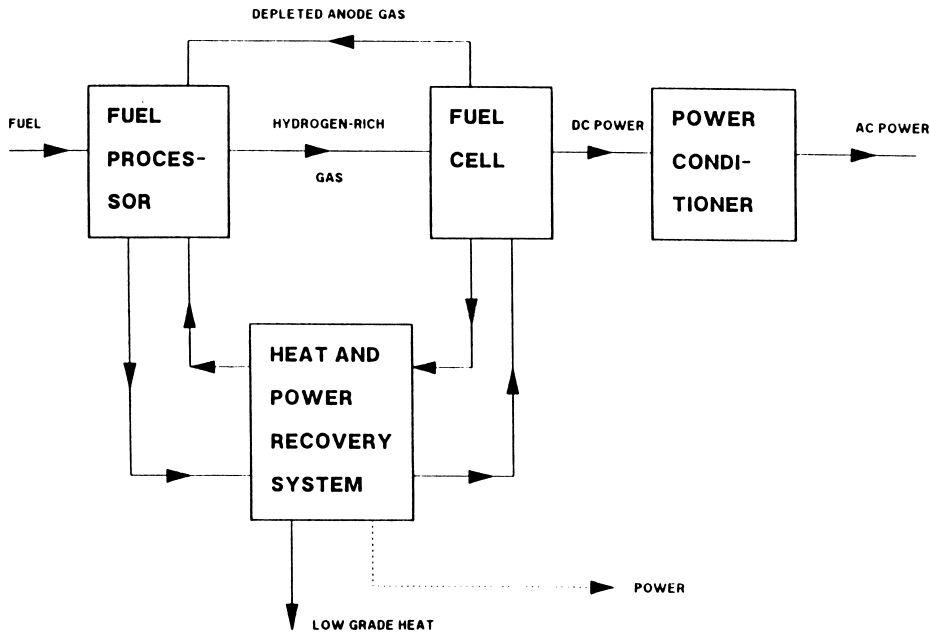


Fig. 4. Block diagram of a fuel cell power plant.

applications (in smaller systems) or additional electricity. Finally, the available low temperature heat can be used for, e.g. central heating applications.

From a system efficiency point of view, system operating pressure is an important parameter. Increased pressure has several beneficial effects on the fuel cell performance because it enables higher reactant partial pressures and consequently higher gas solubility in the electrolyte and mass transfer rates. Besides that, increased system pressure also reduces the size of the ancillary equipment and auxiliary power consumption as long as efficiencies of rotating equipment are high. Since ambient operating systems are less complicated and expensive than their pressurised counterparts, pressurisation looks attractive for systems greater than 1 MW_e.

5. State-of-the-art

The AFC has already been used extensively in space technology and for some special military applications, e.g. submarines and armoured vehicles. It is possible that AFCs will be used for civil purposes in the future, e.g. for transport.

A disadvantage of the alkaline cell is its sensitivity to carbon dioxide. The fuel and the oxidising agent usually contain carbon dioxide which reacts with the alkaline solution, producing carbonates. The fuel must therefore be very pure hydrogen and the air supply containing the oxygen must be free from carbon dioxide. Another disadvantage is the high cost. Extensive research work has still to be carried out in order to construct an economically viable fuel cell for civil purposes. Concerning the long-term energy future (hydrogen economy) AFCs offer a rather mature technological solution and the extent of their application will depend on the relative progress of other competing fuel cell systems such as the PEFC.

The PEFC is the most recent development in fuel cells. The PEFC uses proton conducting polymeric membranes as an electrolyte and as such offers the following advantages: (i) no corrosion problems, (ii) simple fabrication, (iii) CO₂ tolerance, (iv) operability with hydrogen and reformed fuels, (v) possibilities for high power densities. This cell is most suited for small units as used in the transport sector, where fuel efficiency is 2.5–3 times lower than for fuel cell systems and pollution control is still a problem, especially in urban areas.

Like the conventional combustion engine, it can be used at low temperatures: it heats itself to a working temperature of about 80–100°C. It has been successfully used in space technology. Key barriers for commercial applications are: (i) the electrodes are made of platinum or platinum compounds, (ii) poisoning effects of carbon monoxide (contained in reformed fuels), (iii) water management problems and (iv) costs. However, encouraging R&D results have been recently achieved in overcoming these

barriers. Platinum use has been reduced drastically and the problem of the poisoning effects of the CO has been tackled by selective catalytic oxidation, while cost reductions and essential power density increases have been demonstrated.

Based on these recent developments PEFC technology was selected by the US Department of Energy (DOE) as the technology for mid-term introduction of fuel cells in light duty applications (National Programme Plan for Fuel Cell in Transport). DOE studies have indicated prices in the range of US\$50/kW_e to be possible in case of mass production. Another promising aspect of the PEFC is the possibility to be directly fuelled by methanol.

Many of the major car manufacturers have embarked on dedicated fuel cell car development programmes, either or not in co-operation with fuel cell developers. First and second generation prototypes have been presented and small fleets of buses are being incorporated in city traffic, commercialisation is expected in about 7–10 years.

In PAFCs, phosphoric acid is used as the electrolyte. Phosphoric acid is a stable acid, but reacts aggressively with base metals. Consequently, only platinum or platinum alloys can be used for the construction of the electrodes. The cell has the important advantage that fuel based on coal gas or natural gas can be used without the danger of poisoning the electrolyte, but CO has to be converted/removed to avoid platinum poisoning.

PAFC stacks have been under development for about 30 years, and are considered as the most mature type of fuel cells. PAFC technology is presently at the initial phase of commercialisation at the level of 200 kW_e. The promising competitive advantages are generally expected in the small capacity range for dispersed/on-site power generation in remote and urban areas. Today, world-wide, 85 fully packaged 200-kW_e PAFC systems are in operation. In Milan, a 1.3-MW_e installation has been in operation since 1992; in Japan, an 11-MW_e unit has been in use since the end of 1991 and 1- and 5-MW_e commercial prototypes are being tested. With these projects, the practical application of this type of fuel cell has become a fact. The ultimate achievable net electrical efficiency of natural gas fuelled PAFC systems is limited to about 47% LHV. This is expected to be achieved by pressurised operation. Hydrogen fuelled PAFCs are expected to achieve ultimate efficiency goals of about 60% (LHV).

The present capital cost of PAFCs is in the range of US\$2500–5000/kW_e. However, according to recent information from industrial sources there are now substantial improvements towards capital costs of US\$1500–2000/kW_e for the 200-kW class in the near term. Due to the low operating temperature (<200°C), the opportunities for enhancing the efficiency by adding a bottoming cycle are limited, making PAFCs unattractive for large scale power production, limiting their application to district heating and low temperature industrial combined heat and power (CHP).

The MCFC differs from the three cells mentioned above in that the use of molten carbonates results in a much higher operating temperature of around 650°C. The electrolyte is very corrosive and consequently high grade construction materials must be used. Due to its operating temperature nickel (anode) and nickel oxide (cathode) electrodes can be used.

A fuel gas derived from fossil fuels contains CO₂ and CO. The MCFC is both insensitive to CO₂ and, with the nickel/nickel oxide electrodes, immune to poisoning by CO. With the operating conditions in the MCFC, CO is oxidised via the water-gas shift reaction to CO₂ with the production of hydrogen. In the MCFC the oxidising agent for hydrogen are carbonate ions. These ions are formed at the cathode. Therefore, the oxidant gas must contain CO₂. In practice, the CO₂ is provided by recycling the anode off-gas to the cathode.

The two major designs of MCFC are external reforming and internal reforming. Recently, field tests of a 2-MW_e internal reforming system at the city of Santa Clara and a 250-kW_e external reforming system by San Diego Gas and Electric, both in California, have been performed and a 280-kW_e system was started up in Germany. These will be followed in 1998 by a 1-MW_e system in Kawagoe, Japan. Extensive development is still required before commercial applications come within reach. Commercially competitive multi-megawatt systems may, therefore, not be expected before the year 2005.

The SOFC is based on a solid electrolyte. It operates at a temperature of 1000°C. At that temperature, natural gas can be easily converted into hydrogen such that pre-processing (reforming) of the fuel gas is unnecessary and conversion can take place in the cell. The high temperature heat released during the process can be used as process heat for high grade applications. Further research work is needed to improve construction materials and methods of manufacture. The solid oxide fuel cell has been developed to a capacity of 100 kW_e. Commercially competitive multi-megawatt SOFC systems are not expected before the year 2005–2010.

Both high temperature fuel cell types (MCFC and SOFC) offer the highest achievable electrical efficiencies related to the system configuration. These fuel cell types reject their heat at temperatures suitable for raising high quality steam. It is, therefore, very promising to integrate them with a gas and/or steam turbine bottoming cycle. MCFC and SOFC systems fuelled with natural gas are an attractive option for distributed power and CHP applications in the capacity range of 2–20 MW_e. For these systems, developers envisage a sharp decrease in capital costs from US\$1800/kW_e in the year 2000 to US\$750–1000/kW_e by the year 2010. Large scale power generation with high temperature fuel cells is not envisaged before 2010. For large scale power generation (>200 MW_e) both the MCFC and SOFC can be fuelled with natural gas or with coal/biomass derived gases. Due to the operating temperatures the integration with steam turbines (in the case of MCFCs) or integration with gas and steam

turbines (in the case of SOFCs) is feasible. For natural gas fuelled MCFC or SOFC systems net efficiencies in the range of 65–70% LHV have been predicted. For coal fuelled systems efficiencies up to 55% LHV are anticipated. Long-term capital costs are estimated to be in the range of US\$650–850/kW_e for the natural gas fuelled systems and in the range of US\$1600–1800/kW_e for the coal fuelled systems.

6. Markets and prospects

Based on the characteristics of the different types of fuel cells, all possible power generation applications can be envisaged. In the near future the most promising stationary application will be decentralised co-generation, for industrial use as well as for houses and buildings in urban areas. The fuel cell systems will be installed at the location of the end user, where the produced heat and power can be used on site. The generated heat may be used for tap water, central heating, steam production or absorption cooling. For individual use, mini and micro CHP systems in the range down to 1–10 kW_e are being investigated.

Fuel cell systems for co-generation in urban areas, for use in offices, restaurants, hotels, hospitals and housing estates, should have a capacity up to approximately 1000 kW_e. Urbanisation is characterised by a high heat to power ratio in energy demand. Given the low heat to power ratio of fuel cell systems with respect to the demand in this market segment, the utility sector will be the obvious partner to realise and operate decentralised fuel cell CHP projects. For application in the industrial sector fuel cells in the multi-megawatt range are needed. Possible users are to be found in (petro)chemistry, the food industry and the (heavy) steel industry. For industrial application the temperature level of the produced heat will be a significant parameter for market penetration.

In various branches of industry large amounts of hydrogen are available, which makes the application of fuel cell systems an attractive option from both an efficiency and a cost perspective. In addition, industry seems a very appropriate sector to start the introduction of fuel cell systems, bearing in mind the experience in this sector with process units similar to fuel cell systems.

For stationary applications, the objective of most manufacturers seems to be the development and supply of 2–50 MW_e systems to meet the demand for more decentralised capacity. In addition, the fuel cell may be used to increase the existing capacity for central electricity production. The fact that fuel cells keep their efficiency under partial load makes these systems also very suitable for load-following operation. For this a controllability of the reformer of approximately 10%/min is needed. The use of fuel cells for peak load is also possible, but in that case application of gas turbines would be an important alternative because of the lower installation costs and ‘fast’ characteristics of gas turbines.

The future market for fuel cell systems will mainly be dictated by the demands for low cost, high efficiency and low emissions. With respect to emissions, fuel cell systems have definite advantages. NO_x emissions are very low, SO_2 emissions are almost zero and CO_2 emissions are directly related to the efficiency and the primary fuel used. Fuel cells, however, have to face (severe) competition of still developing technologies like high efficient gas turbines in combined cycles, which will also be able to achieve high efficiencies. The role fuel cells will play in the future will be set by the economical efficiency and emission regulations. Most important applications, therefore, are expected where avoiding pollution is a crucial side-effect like in densely populated areas. Market penetration will depend on the price for the total system offered compared with prices for the competing technologies, equipped with emission control equipment.

This is true for both stationary and mobile applications. Developers of mobile systems aim at the 20–250 kW_e range, usually with support from batteries for peak power. An especially important parameter in the transport application is the fuel infrastructure. On this point consensus has still to be established, which is also reflected in different approaches: on-board storage of hydrogen, on-board processing of a variety of fuels or direct feed of methanol to a modified PEFC.

Crucial for the market penetration of fuel cell systems are specific investment costs and operating costs. The specific investment costs will be determined by the potential sales and hence the price restrictions associated with mass production. It is, however, important to realise that the permissible (competitive) specific investment costs for fuel cell

systems are different for each country as parameters affecting these specific investment costs may differ considerably. Besides that, the permissible specific investment costs will also vary with the type of application.

7. Conclusions

Fuel cells are a relatively new but fast developing energy conversion technology with a unique combination of attractive characteristics. Fuel cell systems can be designed for any application in which the production of power or power plus heat is required. Their future market will be mainly dictated by the demand for low cost production of these commodities in combination with environmental regulations. Superior power and emission characteristics have been demonstrated but specific investment costs and long term operation and maintenance costs have still to be determined. However, their considerable development potential in comparison with existing and competing technologies, and the transition from low volume/high cost to high volume/low cost production can make fuel cells a serious contender for the title of this paper.

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