

Abstract

This article describes the recent developments of PEMFC technology with an interdisciplinary approach. Although Fuel Cells are known for clean and reliable electricity production, recent advances of polymer electrolyte technology have fired the interest of both automotive and political sectors ; this is due to the belief that PEMFC may become the key technology for a sustainable transportation system, besides a state-of-the-art solution for portable and stationary power and heat production. The technical analysis of the fuel system embraces different fuel strategies to help understanding the complex interactions between the system complexity/efficiency, present energy/fuel-supply infrastructure and long-term hydrogen/renewable patterns. The cost of electricity of two PEMFC systems has been evaluated from conservative investment costs assumptions. The results show that although the cost per kWh is still quite high (0.26\$Fr/kWh), no major technical barrier are foreseen to slow down PEMFC technology from a fast advancing on its learning curve once industrial production has begun.

Introduction

The problems of toxic emission and resource depletion caused by economic activity are leading to technology changes which will affect the world industry energetically and logistically. Although the geopolitical issue of long term fuel availability is no more on the priority agenda, in favour of greenhouse gases concern, with the associated initiatives to increase efficiencies of both power and automotive industry, strategically, the several regional conflicts, and the gradual establishment of democratic regimes in developing world suggest a more conscious use of fossil fuels in the future. This may be due either to limited availability or from a shift of revenues toward the producing countries. In parallel, mature economies are endorsing sound environmental policies to increase the use of renewables and limitate the damages of massive IC engines utilisation, notably in urban centers. These figures give the impression that a new and more active role for governments in the energy and transportation is at the forefront. If the power industry - traditionally public-owned - has gradually lowered the dependance from oil by diversifying its fuels (first nuclear energy, now gas and renewable sources), and is now undergoing structural changes enhanced by legislative de-regulation and stringent emissions and efficiency targets, the car industry is albeit much more oil-captive and efforts to reduce the emissions - the major driver of research for alternative vehicles – (GPL, NG, hybrid and electric) have not led yet to any relevant market implementation. On the other hand, concern in the public is leading to the establishment of programs for dramatic emission reduction, whose most famous exemple is the California Air Resource Board (CARB) mandate, requiring Zero Emission Vehicles (ZEV) in the coming years. Fuel cells have been called “the Clean Machine” or “Engines of Change”² or, more prosaically clean, efficient power generator, with no moving parts, but little matters : if the fuel cell was invented more than 150 years ago, with early electricity developments, today the FC development focuses on combined heat and power systems and FC vehicles (FCV). The article analyses how the “gaseous battery” has evolved, with comparison to traditional power systems – internal combustion engines (ICE) and gas turbines (GT) - by the cost and efficiency figures. Different gas chemistry schemes to produce the fuel have been included to cover different routes for commercialisation and provide elements for further research. The study performed three tasks :

- to provide a literature review about hydrogen energy development and, more specifically, assess the PEMFC technology status ;
- to analyse technically the PEMFC systems with an interdisciplinary approach and identify the major problem : technical integration and public acceptance ;
- to estimate economically the major subsystem components for a technology in a pre-commercialisation stage and quantify the expected investment and electricity cost for two PEMFC systems.

PEMFC Technology

Fuel Cells (FC) are electrochemical devices that allow the direct conversion of an oxidisable fuel into electricity, with an electric efficiency - calculated from the Lower Heating Value³ (LHV) of the fuel - ranging from 40 to 60%. A FC stack consists of many modules in electrical series. In such a configuration the electric current delivered will be that produced by each element, but the total voltage will be the sum of each module's. The Bipolar Plates accomplish the

¹ This article summarizes the Master Thesis for Cycle PostGrade en Energie at the Swiss institute of Technology of Losanne, 1997.

² See R. Williams « The Clean Machine » and G.E.H. Ballard contribution (« Engines of Change ») at the XI Hydrogen Energy Conference (HEC), Stuttgart, October 1996.

³ The heating value of an oxidisable substance is equal to the recoverable thermal energy resulting from the complete combustion ; when the the water produced from the hydrogen oxidation is condensed we talk of higher heating value (HHV), otherwise it is the lower one. Efficiencies are often calculated from the LHV, although the PEMFC produces liquid water and the HHV is applicable.

interconnection element between MEA's with the anode of one cell, being in electrical contact with the cathode of the following one, while maintaining separation of reactants. The design of the bipolar plate is of major importance for cell efficiency and durability : at the anode fine gas distribution, current collection, membrane humidification and heat transfer have to be preserved, while at the cathode water removal and air delivery add complexity. In the field of stack engineering the main problem has been ensuring a series of FC modules in a compact frame by means of cheaply-produced BP. The stacks of different FC companies can be distinguished by the design of the bipolar plates. Ballard started with graphite-machined bipolar plates but now produces metallic-grooved units. It has been proven that a cross flow geometry in gas distribution improves reaction speed, but to ease water removal vertical grooves are more suited. De Nora has chosen a design, which might be better, suited for straightforward mass production. Their BP design is characterised by a gasket matrix where electrode and collector are fitted. The collector, derived from electrolysis technology, is metallic and it can be produced with inexpensive techniques. Siemens' design involves corrugated metal sheets with an internal cooling fluid, whose manufacturing is done by stamping techniques⁴. Ballard accomplished major progresses by replacing the DuPont membrane with a new, high conductive membrane from Dow Chemicals (XUS), but costs pushed the company to develop its own partially-fluorinated polymer. Power density improved of a factor 25 (>1kW/lit). De Nora stacks have gone from 0.1 to 0.3 kW/Lt; air-cooled stacks and pressurised operation are expected to increase these values.

Fuel Cell Electrolyte	Temp (°C)	Status	Disadvantages
Polymer membrane (Nafion™, Dow, Asahi, Gore, PSI, Dais)	70	Mobile and stationary applications (0.1 – 300 kW) system assessment	CO < 20 ppm
Alkaline (85 wt% KOH in water)	80-150	Mature technology for air independent applications (1-40 kW)	Pure H ₂ and O ₂ ,
Phosphoric Acid	200	Commercial 200kW CHP units (300\$(kW), portable 200W)	η_{el} max 43% (nat. gas), price
Molten Carbonate (Li ₂ CO ₃ + K ₂ CO ₃)	700	Field testing (1MW)	Corrosion, limited modularity, low η_{el} Need CO recycling
Solid Oxide (ZrO ₂ + YO ₃)	800-1000	Pre-commercial (3-10kW) 200kW tests	Max 0.25 A/cm ² at 0.7 Volts
Direct Methanol (Nafion™)	90-130	Laboratory (few W)	Hardware stability High Pt loading, lifetime
Heteropoly Acid PWA (H ₃ PW ₁₂ O ₄₀ ● 29H ₂ O)	20-50	Laboratory (few W)	Dissolution of the electrolyte

Table 1 - Types of fuel cells and their status

Applications

The electricity generation industry is mainly interested in overall system efficiency and lifetime although now emissions abatement has become a major concern as well. FC systems generating hydrogen from fossil fuels are noiseless and have practically no air polluting emissions (particulate, NO_x, SO₂). The greenhouse gases (CH₄, CO₂) emitted, determined by the overall system efficiency (40%) and the anode exhaust, place the FC system above small gas turbines (< MW units), while, on the other hand, the comparison with engines is less favourable, since diesel and gas unit have electrical efficiencies up to 44% and 39% respectively. Saying this, FC systems do have comparative technological advantages because they are noise and air pollution-free without clean-up devices, which reduces operation & maintenance costs, while the absence of moving parts increases the lifetime to about 40.000 hrs. These features allow FC to be user-sited even in densely-populated areas, thus avoiding T&D lines cost and the increasingly important visual impact. Concerning automotive applications, PEMFC system must fulfil very strict performance and cost requirements, besides a severe problem of distribution infrastructure when fuels other than hydrocarbon are used. The FCV shares some attributes with the EV (modularity, perhaps similar manufacturing processes, need for electric drivetrain, etc.) while its fuel storage and high temperature circuits are comparable with IC vehicles. A special attention in the field of mobile FC systems has to be given to the all-round storage/stack safety. Comparing to the stationary applications, efficiency and lifetime will be partially relaxed (a lifetime of 5000 hours is acceptable) but the cost of the FC powertrain has to compete with car engines. Economic and technical targets of mobile PEM technology are an investment cost of approximately 30\$/kW (stack) and power density of 0.4kW/kg⁵. It has to be noticed how research has recently shifted from the stack power density to the overall system definition and integration, including fuel processing, infrastructure, public acceptance and set up of mass-manufacturing equipment to compete with conventional

⁴ E. Grecksch & T. Moser : « PEM Fuel Cells : Development and Commercialisation », Intertech Conference, Chicago, september 1996.

⁵ These are the targets of the U.S. program Partnership for a New Generation of Vehicle (PNGV) in ref. 35 above page 36.

vehicles. Besides the aspects of volume, weight and vehicle range, weight distribution, transient response and security in accident or fault situations are of major relevance for customer acceptance.

Fuels and Balance of Plant

In the Economics of Hydrogen fuel, the most economical hydrogen generating process for medium and large plants is Methane Steam Reforming (MSR). Small production (10's kW) units use Partial Oxidation (POX) or Autothermal processors (ATR), suited for « pedal responsive » applications in automobiles. Processes as electrolysis and pyrolysis of biomass, to produce methanol, can be profitably integrated with renewable energies and, having no CO₂ release on the whole fuel cycle, show an advantage in the long-term perspective of a non-fossil based economy, with positive applications in the electrification of developing countries. A list of comparative hydrogen production costs is given below.

PROCESS	READINESS	COST (\$/GJ)	EFFICIENCY (%)	
			ENERGY	EXERGY
Methane Steam Reforming	Mature	5	86	78
Partial Oxidation of oil	Mature	9		
Coal Gasification	Mature	10	59	49
Hydroelectric electrolysis	Mature	10-12	85	
Wind electrolysis	R&D/mature	30	NA	
Thermochem. Water Decomp.	early R&D	30-50	21	19
Solar PV Electrolysis	R&D/mature	30-50	8 (from state-of-the-art)	
Biomass gasification	R&D/mature	12	NA	
New...photoelectrolysis etc.	Laboratory	NA	NA	

Table 2 - Hydrogen generating processes⁶

Concerning the key BOP components, the efficiencies of different inverter technologies are quoted: Mosfet and GTO technology efficiencies decrease at part-load, the software-controlled Insulated Gate Bi-polar Transistor technology can maintain > 90% over the load range.

TECHNOLOGY	CURRENT STATUS	TARGET	Cost(\$/kW)
MOSFET Efficiency	82-86%	95%	N/A
GTO Efficiency	86-90%	95%	N/A
IGBT Efficiency	95%	-	30

Table 3 - Inverter technologies⁷

In methane steam reforming a pressurised mix of sulphur-free methane and super-heated steam react over a nickel-based catalyst in a tubular reactor. This is generally followed by two adiabatic water/gas shift reactors to convert CO to CO₂ with extra steam and produce more hydrogen. Half of the hydrogen is produced from the water. SR process have a high yield with a high steam to carbon ratio (3-4) but it features a poor transient response since whenever a higher flow of hydrogen is demanded the resulting higher reformer heat demand comes before extra anode off gas is available to be burnt. The maintenance cost of the heat-exchange tubes and a general bulkiness of the system sometimes offset the simplicity of operation and good yield. The exothermic nature of the partial oxidation reaction allows a decrease in reactor size, costs and maintenance for on-board fuel conversion of FCV. In searching a synthesis between exothermic and endothermic reactions, research is oriented toward an internally heat-balanced reforming process : the autothermal reformer. In ATR the *in situ* oxidation zone of the reactor supplies the heat needed by the endothermic steam reforming reaction, which takes place in the same chamber downstream. ATR, are compact with good transient response benefiting of both SR high yield (efficiency) and POX capability to treat different hydrocarbons (LPG, diesel or gasoline). One additional advantages of the ATR is that the sulphur compounds do not damage the catalyst at temperatures above 800°C. A comparative analysis shows methanol SR as the most efficient fuel processor, with methane and ethanol SR following, while ATR and POX have lower values, but the comparison of fuel processor efficiencies may be confusing because it does not account for the energy inputs other than the fuel, as the heat recovery from the anode exhaust gas. It is thus recommended to look the overall system efficiency reflecting not only the reformer technology, but, mostly, the effectiveness of thermal integration of the system. The autothermal process, with

⁶ Sources : H. Audus et al. : « Decarbonisation of Fossil Fuels : Hydrogen as an Energy Carrier », Proc. XI HEC, Stuttgart 1996. Cited from U.S. DOE : « Hydrogen programme implementation plan » (1990). M.A. Rosen for the efficiencies (XI HEC proceedings, 1996).

⁷ Source : Prof. A. Rufer, LEI, EPFL.

conventional fuels as LPG, NG or reformulated gasoline, may become an essential element for mobile applications of PEMFC. Platinum electrocatalysts have a CO tolerance between 10 and 20 parts per million volume (ppmv) of dry gas fed to the anode and optimised reformers can at best achieve 0.5% CO content : a clean-up system is needed before the reformed gas enters the FC. We distinguished between separation processes (Adsorption and Membrane processes) and Chemical Conversion Processes (water/gas shift conversion, selective methanation, selective oxidation). PROX reactions go back to WW1 when gas masks used by the army (to oxidise the poisonous CO) had iodic acid in a sulphuric solution, which impregnated powdered pumice ; the iodine and CO₂ produced were retained by sodium carbonate. Hopcalite, a mix of Mg, Cu, Co oxides and Ag was subsequently preferred, the main problem being that the catalyst functions on a dry gas only. Early work⁸ on integrated FC systems was based on the assumption that the Hopcalite catalyst of the selective oxidation reactor is effective for the operating environment. Moreover optimum dispersion of the precious metal catalyst (leading to high activity) we distinguished three key parameters: operating temperature; residence time; air addition rate and mixing to match the CO concentration ($\lambda \approx 2$). The first two affect both catalyst selectivity and conversion rate, whilst the third concerns the selectivity only since excess oxygen leads to hydrogen burn up. At temperatures higher than 200° C more CO appears, because of the reverse water /gas shift reaction. One of the main problems is the absence of a real time CO sensor to make system response to CO concentration fast enough. A control strategy, where the difference in H₂ content between inlet and outlet is controlled, varies the amount of injected air on a real-time basis, in order to continuously maintain the desired oxygen-to-carbon monoxide ratio (2:1), enabling a dynamic response to the power demands on the FC system. The strategy is based on the fact that H₂ content will lower if too much O₂ is injected. The major work on selective reduction gave satisfactory results using a Ni-based CRG-F catalyst developed by British Gas⁹, but these results could only be obtained at precise operating conditions. Toyota methanol FCV employs a methanation reactor with a ruthenium catalyst. Two non-exclusive paths may be :

- I** - Infrastructure for vehicles leads to strong standardisation and rapid economies of scale because integrated mass production conditions create synergies between an alternative fuel and the current energy and industrial infrastructure.
- II** - Dispersed small-scale reformers, peak-shaving electrolysers and storage equipment for stationary FC systems increase gas demand and optimise electricity production, enabling FCV mass production through hydrogen refuelling station.

Markets

PEMFC are suited for the 10 kW - 1 MW powerband where they face a strong competitor : diesel engines can attain 43% (38% with gas) electrical efficiency with a flat behaviour from 40 to 80%¹⁰. Thus, FC need to lower their cost and take advantage of air and noise pollution high value. The market evolution estimations (standby, cogeneration and site supplies) suggest that installed capacity might rise to 45 GW from the actual 20 GW. FC will take around 7 GW. The most promising application is for cogeneration installations, where cost goal is to go lower than 2000 \$/kW. We believe that the share of the power generating market for PEMFC will ultimately be determined by the reliability and cost of the fuel processing and clean up system. The stack will need to operate at high voltage, while size and density relaxation - compared to mobile application - will be an advantage. The market below 10 kW is satisfied by small motor generators and rechargeable-battery sets. Interest for FC comes from the military and aerospace industry and, considering that more than a million small diesel and petrol gensets are sold each year, it is more than a 1000MW market. Uncertainties concern what part of power-only and what CHP will be demanded. By 2020 CHP sales might reach 100.000 units for a combined capacity of 400MW, while electricity generators (1-10kW) will be around 100MW. Strong demand for electricity-only FC may come from the army to replace battery-powered military kit (30-300W), such as night vision systems and communication devices, but the civil customer may emerge from the growing need for reliable power in high value applications : portable computers, home smart metering and communication equipment. 2020 sales estimates for battery replacement vary largely : from 250 to 800 MW. Vehicles demand represents a possible market of 57 GW by 2020, composed for 42GW of FCV and 15GW of commercial vehicles such as vans, buses, trucks and fork-lift trucks. The commercial vehicles are favoured in early commercialisation because they are intensely used, have central refuelling stations and government might take an active role to enhance their introduction (perceived environmental benefit). Other studies foresee 800 units for commercial vehicles by 2010-2015, while FC cars are mass-produced by 2018-2019. The vehicle market will entirely be taken by PEMFC, some applications, as vans and boats, might also present chances for alkaline technology. We evaluated the FC system investment for two stationary hydrogen production schemes : 1) Hydrogen produced in a neighbourhood-scale MSR (100's kW) with a PSA purification unit; electricity is delivered through adapted existing gas lines to the customers, the local utility is the FC owner (5050\$/kW) ; 2) A 50kW ATR/PROX fuel processor producing hydrogen for a private user from gas or liquid hydrocarbon (3400\$/kW). The results of the electricity cost below are derived from the following assumptions:

⁸ U.S. Department of Energy, Contract No. DE-AC02-92-CE50343 : « Multi-Fuel Reformers for Fuel Cells used in Transportation », Phase 1, Final Report, pag. 49.

⁹ Dr. T.A. Smith, ETSU Report F/02/00031/REP : « CO Removal by Methanation for the Solid Polymer Fuel Cell », 1996.

¹⁰ Source LENI, EPFL.

- Insurance cost is 0.3% of investment cost
- Maintenance cost is 2.5% of investment cost,
- Operation cost (personnel) represents two people at 50% (≈ 75.000 SFr.) for MSR and one operator at 30% for ATR technology (22.500sFr.), which has comparatively a lower reliability.

		200kW MSR	50kW ATR
Capital cost	Annuity	236910	45'633
	Annuity/kW	1184.55	912.66
	Hours/y	8322	7'884
	Tot	0.14234	0.11576
Operating costs	Insurance	0.00268565	0.00218
	Personnel	0.04506128	0.05708
	Maintenance	0.09449518	0.01820
	Tot	0.142242	0.07746
Fuel costs	Gas 0.4eff.	0.07500	0.07500
COE SFr./kWh)		0.35958	0.26822

Table 5 - Cost of Electricity for 50 and 200 kW pre-commercial schemes.

Conclusions

Technology exerts a certain fascination nowadays because, intuitively, “there” lies the key to solve a number of different problems. The search for an oil-alternative energy system – started in 1973 to *increase* supply security – has brought new technologies to the market (e.g. nuclear, photovoltaic) and led to improve existing ones, as wind generators or gas turbines. Some technologies based on renewable energy sources are now commercial, but – we know - they can not replace the conventional energy system for both power peaks and transport fuels. Today the Kyoto targets and urban mobility are stringent priorities and the new objective is to *reduce* emissions at the global and local level *to* maintain economic growth. Such a challenge implies a fundamental re-questioning of our development model. Present issues like global warming and transport pollution cast in doubt whether the growing need for energy services can be met with conventional technologies, even with the implementation of end-of-pipe cleaning devices. Thus, it is likely that an improvement of living standard world-wide will require revolutionary changes in both energy production/conversion and end-use technologies. As in the past oil-fired motors did not replace James Watt steam engine because of coal shortage, fuel cells’ future commercialisation will firstly be due to the higher global efficiency – compared to thermal motors – which will make new high-value, environment-friendly applications possible. The following conclusions can be drawn :

- PEMFC stacks have attained the required power density for (zero-emission) vehicle applications;
- Dynamic reformers for hydrocarbons or alcohol’s are available;
- Carbon Monoxide clean-up devices are being tested and optimised to be operated with reformat from (2).
- Renewable electricity will contribute to FC market with decentralised hydrogen generation & storage.

The new paradigm of the information society changes our energy needs and the related structures : from vertical, centralised, production-oriented to consumer-sited, clean and reliable. Concerning transport, improvements in spatial planning, which can reduce distances between activities and lead to less need for mobility, shift the focus from mobility to accessibility. In this sense, the upcoming, interconnected web-shaped society leads to an optimisation of the economic process, accounting full-cycle flows of energy and materials and breaking the barriers to market entry. PEMFC promise to be the cheapest FC technology : metal hardware with plastic membranes and aluminium bipolar plates, shape the silent, non-polluting, ultra-compact energy generator, which makes water as by-product. Although present progresses in the fuel processing do not allow either precise conclusions about the final fuel for PEMFC, or immediate relaxation on the issue of the equipment costs, the significance of the projects under way (and the companies behind them) give us the impression that both mobile and stationary PEMFC systems will not find major technical barriers for commercialisation.