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Technology up date and new strategies on fuel cells

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Abstract

This paper reports the state of art on fuel cells technology, outlines the most significant results reached all over the world and summarises the strategies developed by researchers and producers to get the commercialisation of these systems. In particular, the authors have examined three potential application fields for fuel cells: (i) stationary power plant for electricity production; (ii) portable power applications; (iii) electric vehicles.

The potential market area for each sector of application has been defined and the research activity necessary to overcome the technical problems that are still open have been detailed. The significant projects of main fuel cells producers are also mentioned.

Keywords: Fuel cells; Stationary plants; Portable applications; Transport applications

1. Introduction

At the beginning of the 1960s, alkaline fuel cells were used as electrical generators for Apollo space vehicles: this event can be considered as a milestone that marked the commencement of fuel cell technology development and of its application in civil areas.

Today fuel cells are considered [1] as environmentally friendly and high efficiency systems for the production of electricity, and world wide research efforts have been addressed to the improvement of this technology.

First of all, fuel cells have been investigated as innovative systems able to be integrated with traditional large electrical power plants or to supply electricity as large on-site power generators. For this purpose, in past decades, fuel cell technology development has been addressed to the production of large demonstrative power stacks based on phosphoric acid fuel cell (PAFC), molten carbonate fuel cell (MCFC) and solid oxide fuel cell (SOFC).

Currently, the technical problems encountered in the management of these large power fuel cell plants and their resolution have occurred in the field of specific technologies aimed at the recovery of waste heat (microturbines) and involving projects for the development of small/medium power fuel cell generators intended for a widespread de-regulated electricity market.

The development of a polymeric membrane, with protonic conductivity properties and which is applicable as a solid electrolyte for fuel cells, has been another event that revolutionised the expected applications and opened new sectors with great market potential. The availability of cells based on the solid protonic conductive polymer (PEFC) operating in mild and friendly conditions, without losses of electrolyte and at high power density, has been identified as a real alternative to the internal combustion engine for vehicle propulsion.

The effects on the market produced by a new system of propulsion with high efficiency and low environmental impact are easily appreciated and they justify the great R&D efforts that are being made. The diversification of fuel cell applications also involves, the market of portable generators for civil uses like laptop computers and cellular phones or military ones (back power-packs). In this sector, the driving force for technology improvement involves the potential application of polymer fuel cells with direct supply of methanol (DMFC).

Today, fuel cells have reached a degree of development from which it is possible to envisage future commercial fields where this technology could have a decisive role in many applications where electricity must be produced with high efficiency and low environmental impact. This paper

Abbreviations: APU, auxiliary power unit; BOP, balance of plant; DFCV, direct fuel cell vehicle; DMFC, direct methanol fuel cell; FC, fuel cell; FCV, fuel cell vehicle; FP, fuel processor; ICE, internal combustion engine; MEA, membrane electrode assembly; MCFC, molten carbonate fuel cell; PAFC, phosphoric acid fuel cell; PEFC, solid polymer fuel cell; PEM, proton exchange membrane; PFCV, processed fuel cell vehicle; PNVG, partnership for a new generation vehicle; SOFC, solid oxide fuel cell; UPS, uninterruptible power service

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reports the main aspects of an analysis of fuel cell technology and summarises the world state-of-the-art, critical factors and strategic goals.

2. Fuel cell stationary applications

During the last decade, very significant structural changes have been observed in the field of electricity production and distribution. This evolution seems to be just beginning and new and more profound modifications can be expected. These changes in energy policy are mainly due to two different factors: the de-regulation of the electricity market and the Kyoto commitments.

De-regulation is already a fact in the USA, and now is proposed in Europe. Its effect is intended to be the disappearance of monopoly regimes, a more flexible electricity market, a decentralised energy production and a lowering in the price of electricity, and it strongly influences the development of new technology; in fact, the producers are encouraged to promote the installation of plants that need shorter construction time, require less risk capital and offer faster investment payback [2]. Thus, in the next decades electricity production will be addressed preferentially towards high efficiency, dispersed, small power plants at the expense of existing large power plants. Strong efforts will be made to develop new technologies, like fuel cells, that fulfil ambient safeguard commitments and the need to improve production efficiency.

Fuel cell technology is a favourable candidate for the development of stationary plant for several reasons, such as low environmental impact, high electric conversion efficiency (up to 50–55%) independent of size, production of heat usable for co-generation cycles, integration with gas turbine and fuel flexibility. Despite their high potential, fuel cells are not ready for full commercial application for stationary plants because of their high cost and limited durability. More research efforts are needed to meet the cost of 1000 Euro/kW and life time of 40,000 h that are EC targets for 2005 [3]. A survey of the electricity production market for possible fuel cell applications shows some niche applications, which can be grouped (Table 1) as; small power plants for residential application (less than 10 kW), medium/large power plants for industrial and commercial use (from 10 to 300 kW) and large power plants (up to 20 MW).

Table 1
Power range and field of application for FC stationary plant

	Electrical power	Field of application
Small power	1–5 kW	Micropower — domestic
	5–10 kW	Domestic — residential
Medium power	10–100 kW	Residential
	50–300 kW	Commercial
Large power	250 kW–10 MW	Power station

2.1. Small power plants

The field of small power plants mainly concerns applications for domestic use. In this area, customer needs do not fit very well with fuel cell system characteristics. First of all, the energy requirement of a household as a function of quality (electricity or heat) and time (variations during the day and during the seasons) is not very suitable as an optimal application of a fuel cell. Market analyses showed that an average domestic application present yearly ratio between required heat and electricity equal to 5.45, while for a fuel cell the optimum value for this parameter is in the order of unity. Furthermore, variations of this ratio are produced by the fact that the electricity demand has two peaks per day and the heat requirements change with the external temperature. Thus, a suitable application in the domestic field needs an integration of the fuel cell system with an external grid. This opportunity is not allowed when a monopoly regime manages the sector of electricity production.

Today, the greater flexibility reached by fuel cell devices together with the market liberalisation of electricity production has raised the interest of producers in developing small power fuel cell systems for these applications, mainly supplied by natural gas that is favoured by the existing grids.

The main development plans for applications in this field point to PEFC technology due to its simplicity and ability to be assembled readily by industrial processes. Thus, Sanyo is developing a 2–3 kW class co-generation system fuelled by natural gas [4], Plug Power has formed a joint venture with the Power Division of General Electric to develop PEFC systems for domestic uses and Vaillant, in co-operation with Plug Power, is developing a program on small scaled PEFC for heating purposes with a production target of 30,000 units in Europe by the next 5 years [2].

The commercialisation of small power fuel cells is not strictly related to the domestic field but attempts have been made to find new potential markets such as electric generators for users requiring stable, reliable and high quality energy, like hospitals, computer centres, uninterruptible power service (UPS) and remote applications.

The commercialisation of small power fuel cell generators seems to be favoured if it is based on a SOFC core module that can release high quality electricity at high efficiency. Siemens–Westinghouse [5] and Sulzer/Hexis are engaged in two programmes based on the commercialisation of 5 kW and 3–5 kW SOFC prototypes, respectively.

UPS applications based on the use of a fuel cell as a secure power source are one of the most interesting among the new applications. The scheme of a fuel cell-based UPS can be as shown in Fig. 1. This scheme includes dc electricity production from a fuel cell which is directly connected to a conventional UPS; the surplus of electricity produced by the cell can be supplied to the grid and the emergency batteries can be reduced in capacity.

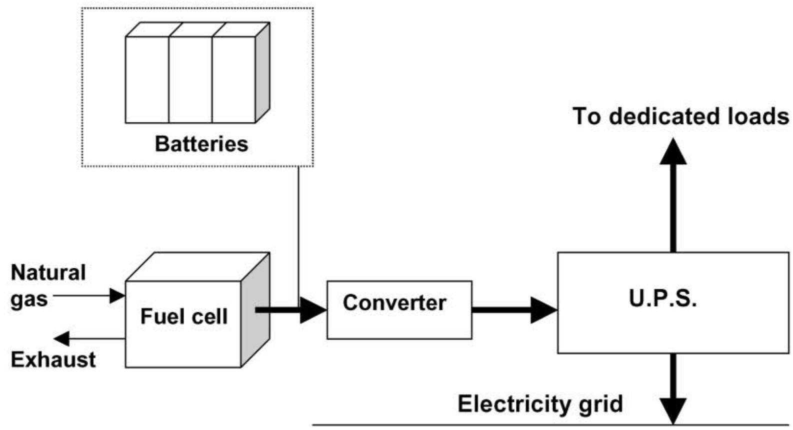

Fig. 1. Scheme of a potential UPS device based on fuel cell technology.

2.2. Medium/large power plants

Medium/large power plants appear to be the optimal size for fuel cells. There are several reasons for this, and the most important of these are: (i) the energy user requirements can be fully satisfied by fuel cells; (ii) for these applications different types of fuel cells (PAFC, PEFC, MCFC and SOFC) can be considered; (iii) the experience gained with several applications indicates that the technology is sufficiently reliable for the development of fuel cells up to 200 kW; (iv) high performance co-generation plants can be designed as well as plants integrated with microturbines; (v) natural gas from an existing grid can be used as fuel. The main applications in this field concern plants for electricity production and co-generation for buildings, and for industrial and commercial applications. In parallel to the above listed advantages, other significant features of these plants are the production of premium quality power, the possibility of using an independent source that can operate for ‘‘on-site’’ production or as a continuous power backup or as an uninterrupted power supply (UPS).

The nominal power of the planned plants ranges widely from a few tens to some hundreds of kW, even if the established trend is to develop stack modules of limited power (not bigger than 250 kW) and to exploit the modularity of these systems to connect them in large power scaled plants.

The first commercial experience of fuel cell site plants has been based on the application of 200 kW powered PC-25 PAFC units. The IFC (a division of United Technology Corporation), with Toshiba Corporation and Ansaldo S.p.a. as partners, developed this plant which shows a performance of 40% LHV, which can reach 80% with co-generation [6].

Basically, the PAFC results in an attractive system due to its good electrical efficiency and an operating temperature (200°C) that makes convenient the recovering of heat and reduces the risk of CO poisoning. Experience has shown that the costs for these plants are about US$ 3000 per installed kW; this appears to be a severe limitation for the wide spread use of PAFC. Today, it seems that there is little room available for technology improvements on for further reduction of the fundamental PAFC costs. Somewhat lower costs should be obtained for high volume production by highly automated processes, by improving the electrolyte management, the plant stability and electric control equipment.

For some years, fuel cell producers have regarded PEFC technology with great interest due to features such as easy assembly of cell components, mild operating conditions (temperature from 70 to 90°C), absence of electrolyte losses, high power density (300–900 mW/cm^2), good electrical efficiency (from 40 to 60%) and low capital cost (1500–2500 US$/kW).

In practice, some technological problems remain for the commercialisation of these systems as stationary plants. The first problem is represented by CO poisoning of the Pt catalyst with an adsorption energy that is a reverse function of the temperature, thus, at the typical PEFC temperature (80°C), the adsorption of CO on Pt sites is stronger than that of hydrogen and the result is a drop in the cell performance [7]. Two possible solutions can be the increase of cell temperature or the reduction of the CO content of the fuel.

Today, the most important goals for PEFC commercialisation are considered to be

- development of electrodes at low Pt load and a new proton conducting polymer membrane stable at a temperature >100°C;
- optimisation of automated procedures for the production of electrodes and cell assemblies;
- increase of cell operating temperature and improve exhaust gas quality;
- integration between fuel cell and fuel processor subsystem;
- development of CO resistant anode catalysts;
- optimisation of CO selective oxidation processes.

Producers regard high temperature fuel cell technology (MCFC and SOFC) as a good candidate for medium/large scale stationary power plants. Thus, a large number of prototypes or demonstration plants have been assembled and tested around the world. Different research companies from the USA, Japan or Europe are strongly involved in the development of stationary power plants based on molten carbonate or solid oxide fuel cell technology. The reasons for this strong interest are the favourable temperature that allows an appreciable integration with co-generation cycles, full tolerance to different fuels and the possibility to develop the internal reforming concept.

In the last decade, MCFC stationary plants, have probably been the most significant type and 100/250 kW scale stack operations have been successfully demonstrated. The experience gained in the laboratories and plants has also revealed some basic problems [8], such as cathode dissolution, hardware corrosion, electrolyte losses and low plant compactness, all of which need to be solved. Today, MCFC commercialisation has been planned in the USA, Japan and Europe [9–11], with strategic programs devoted to obtain higher performance and endurance through: (i) improvement of the electrolyte and electrolyte support; (ii) reduction of electrolyte losses; (iii) increase of operating pressure; and (iv) development of compact configurations that closely integrate the stack to BOP subsystems.

Other research activities have been initiated on recycling of on material used in stacks and on the investigation of the use of coal-derived gas.

SOFC technology offers systems with good power density (300 mW/cm^2), high operating temperature (1000–800°C), good fuel flexibility and very high electrical performance mainly if integrated with gas microturbines. For the above reasons, most active fuel cell technology developers believe that the commercialisation of SOFC systems can be an obtainable target. For example, the NETL, National Energy Technology Laboratory of Department of Energy (DOE) promotes a development program that intends to commercialise SOFC plants, in 2003, at a cost of 1000–1500 US$/kW and an efficiency from 50 to 60%, and to achieve, by the year 2015, 80% efficiency, no pollutant emissions, and 40,000 h of life time at a price of 400 US$/kW [6].

The spread of generators based on SOFC technology can be successful but some strategic technical aspects have to be solved. Development of a low cost technology for the production of cheaper and durable cells is a critical issue. Advanced materials that lower operating temperature and then reduce interfacial ageing processes, stack technology based on planar supported electrode configurations and nanosized starting materials are the principal research topics that should be developed.

SOFC/microturbine combined cycle systems are considered to be a great challenge in the field of high efficiency plants for electricity production (efficiency >70%). The development of these plants critically requires the production of heat exchangers (that integrate cell and turbine) whose material resist at $T > 700°C$, the reduction of the cost in volume production, low cost manufacturing and optimum integration between major components.

2.3. Large power plants

In the field of large dispersed plants (1–10 MW) fuelled by natural gas, both PAFC and MCFC stationary plants have been set up and further development can be proposed based on the experiences carried out during two decades of tests.

The first prototypes of MW scale power plants installed in the world (1982) have been the 4.5 MW New York plant (by Cons. Edison) and the 4.5 MW Tokyo Electric Power plant. Both plants have been based on PAFC technology. More recently, Chube Electric power Company installed an 1 MW MCFC plant in Kawagoe and Energy Research Corporation (ERC) realised a 2 MW plant demonstration in Santa Clara (1996). This was formed from 16 atmospheric pressure, internal reforming MCFC stacks of 125 kW and achieved an efficiency of 44% LHV. These experiences and the severe technological problems encountered have supplied some important information to guide the further development of large scale power plants. Today the producers are oriented toward the construction of MW scale power plant based on the development of compact single modules of a few hundred kW (in the order of 250 kW) instead of very big stacks whose technology does not seem to guarantee long-term efficient performance.

Further potential applications for fuel cells are: (i) gas-derived coal supplied stationary plants (MW); (ii) repowering of older existing plants.

MCFC and SOFC seem to be the best technology for these applications because of their resistance to the most common fuel contaminants.

2.4. State of art on stationary applications and remarks

Further significant experience with PC-25 PAFC units has been gained by the Gas Utilities — Tokyo Gas, Osaka Gas, Toho Gas and Fuji Electric [12], which since 1989 installed several 50 and 100 kW sized PAFC stacks. Five of these units have accumulated 40,000 h of operation.

Today, several producers are engaged in programs for PEFC commercialisation and Table 2 lists the most significant projects in this field.

Nuvera is proposing three prototype generators of small power. Two of them can be available in 2002 and will involve 1 kW premium modules supplied by hydrogen and propane, respectively. The third is a 5 kW residential module supplied by natural gas and it will be available in 2003.

Actually, Ballard G.S. produces plants based on 250 kW powered PEFC fed by natural gas and Toshiba has under development the development of 30 kW class co-generation system for small buildings fuelled by natural gas or propane.

Table 2
Some significant projects for PEFC commercialisation

Producer	Power (kW)	Fuel	Prototype availability	Application
Ballard	1	Processed NG[a]	Yes	Micropower
Ballard	40	–	–	–
Ballard	250	Processed NG[a]	Yes	Commercial
Nuvera	1	Hydrogen	Not yet	Premium
Nuvera	1	Processed propane	2002	Premium
Nuvera	5	Processed NG[a]	Not yet	Residential
H-Power	4/5	Processed NG[a]	Not yet	Domestic
Vaillant-Plug Power	4/6	Processed NG[a]	Yes	Domestic
GE-FCS	4/5	Processed NG[a]	Yes	Domestic
Toshiba	30	NG/propane/biogas	Yes	Residential
Energy	20/50	Processed NG[a]	–	Residential
Fuji Electric	1	Processed NG[a]	Not yet	Domestic
Sanyo	2/3	Processed NG[a]	Yes	Residential
Shatz-Energy	4	Hydrogen	Yes	Remote power

[a] NG: natural gas.

In the field of MCFC (see Table 3), fuel cell energy (FCE) developed, with MTU of Germany, an innovative concept, the ''Hot Module'' where the fuel cell stack is packaged together with hot BOP subsystems. A configuration that closely integrates the stack to BOP subsystems has been selected by Ansaldo Ricerche S.p.a. (I). This configuration will be the basis of the development of a 500 kW class stack. In Japan, development of 250 kW class modules is planned.

SOFC producers are proposing projects whose aim is to commercialise high efficiency generators. Thus, Siemens–Westinghouse focused SOFC on a commercial target offering a hybrid fuel cell/gas turbine system with efficiency up to 60%, and fed by natural gas. NEDO programs foresee the development of a wet processed tubular module that, at 900°C, has a decay rate of 0.3%/1000 h, and planar cells with materials with improved durability, performance and thermal characteristics.

MTI and NREC are developing a plant where a non-pressurised planar SOFC is connected to a microturbine. The system is fed by natural gas and an efficiency (LHV) of 71.2% is expected. Table 3 reports a summary of the most important plans for SOFC commercialisation.

The analyses of experiences gained so far and the direction of new R&D projects on fuel cells seem to indicate that the main fields of application are already outlined but research efforts are needed to reach a good level of commercialisation.

From past experience it appears that it is very difficult to develop large durable power stack modules, while the production of small/medium power plants appears more

Table 3
Some significant projects for MCFC and SOFC commercialisation

Producer	Power (kW)	Fuel[a]	Prototype availability	Application
MCFC				
FCE/MTU	250	NG	Yes	Commercial
	3000	NG	Not yet	Subpower
ANSALDO	100	NG	Yes	Commercial
	500	NG	Not yet	Commercial
HITACHI	250	NG	Yes	Commercial
I.H.I.	250	NG	Yes	Commercial
SOFC				
Siemens–Westinghouse	1000 hybrid	Processed NG	Yes	Subpower
Siemens–Westinghouse	300 hybrid	Processed NG	Yes	Commercial
Siemens–Westinghouse	250	Processed NG	Yes	Commercial
Siemens–Westinghouse	25	Processed NG	Yes	Residential
ZTEK	1/25	–	Yes	Domestic
Sulzer/Hexis	1/5	NG	Yes	Domestic
Sulzer/Hexis	200	NG	Yes	Commercial
Mitsubishi Heavy	5	–	Yes	Domestic
Mitsubishi Heavy	25	–	Yes	Residential
Fuji	1	Hydrogen	Yes	Domestic
SOFCo	1/4	NG	Yes	Domestic
SOFCo	10/50	Diesel	Yes	Residential

[a] NG: natural gas.

promising. Among various types of fuel cells, PAFC represents an established technology that is expensive if compared to traditional power plants. Further, it seems that this technology has little room for improvements that could reduce the investment and maintenance costs.

PEFC will be a preferred candidate to be used for domestic or residential generators. It remains necessary to develop membranes stable at temperature higher than 100°C and to improve the cell resistance to CO.

MCFC has already reached an elevated technological level that has established the development of compact 200–250 kW modules as the right route for commercialisation. The next few years will be crucial for the future of this technology. In fact, either the MCFC plants will be ready to be commercialised and the existing technical problems will be solved, or they will remain at the level of demonstration applications.

SOFC present great potential application, is as integrated generators with very high efficiency. The improving of cell endurance, the development of low cost planar component scale-up and full fuel flexibility are challenge points for the commercialisation of SOFC-based stationary power plants.

3. Portable power applications

The increasing demand for quality, density and time-performance of power supply is the principal driving force in the portable power production market. This particular market includes an increasing number of new products (cassette and mini disc players, laptop computers, cellular phones) with considerable system complexity, and today presents a trend which, after the exponential increase of the last decade, is now reaching a steady state level and a saturated level of competition.

The competition is actually based on the introduction of new products characterised by a high level of customer acceptance; in other words they must be smaller, and lighter, with an increasing number of functions and cheaper than earlier models.

For all these reasons fuel cells are particularly suited as portable power systems since they have good potential in terms of energy density, durability, simplicity of design and low cost. A fuel cell can operate as long the fuel is supplied to the device and this can be easily done from a very small and light tank. Thus, the need for battery recharging is completely eliminated from the system and the life-time of the power source is significantly longer with respect to the present technology.

3.1. Batteries and fuel cells

Fuel cells are currently designed for relatively high temperature (90°C) operation in order to enhance the electro-oxidation kinetics and to increase the ionic conductivity of the polymer membrane; thus development for portable applications, working at room temperature, requires further effort. Comparing the performance of a Li-battery and the present hydrogen or methanol fuel cells at room temperature, one can say that the differences are not very large but are still significant; for example, the Li-battery of a cellular phone can supply 900 mAh (capacity) and 3.6 V operating voltage. The manufacturers generally claim 2 h of operation before recharging under continuous conversation at 0.45 A and 3.6 V or 200 h operation in stand-by at 4.5 mA. In the first case, the power output is 1.62 W and 16.2 mW in the latter case. Considering the typical dimensions of a Li-battery with such a power output, it is thought that the available surface area of, for example, a DMFC stack with same volume and thickness is 50 cm^2, integrated in two planes. Accordingly, the required power density of a FC intended to substitute the Li-battery is about 32 mW/cm^2. This value is about twice the performance that can be achieved with a state-of-the-art DMFC designed for operation at high temperature and pressure, but working at ambient conditions. Thus, considerable effort is needed to develop a FC to be competitive with the Li-battery under such conditions. In order to match the power requirements and dimensions of a portable power system operating at room temperature and atmospheric pressure, a novel system design and novel catalysts must be developed. Considering first the protonic electrolyte, there are few commercial membranes that have suitable ionic conductivity, although further development could open interesting possibilities.

In terms of fuel consumption, the current that must be delivered by the DMFC during operation at maximum power (operation under conversation conditions) is 0.45 A. Considering, for example, the case of 10 cells of 5 cm^2 connected in series which would provide 1.6 W at 0.45 A, i.e. each cell is operating at 0.355 V, the methanol consumption for each cell is 0.21 ml every 2 h of conversation and 2.1 ml for the whole device. Accordingly, a tank of 50 ml of pure methanol will allow about 48 h of continuous conversation or 4800 h in stand-by operation. Consequently, if the customer is used to recharge its battery every 2 days, with a DMFC he will refill his device every 50 days.

3.2. State-of-the-art of portable power applications and remarks

In recent years a major initiative has been to develop new technologies based on non-bipolar DMFC stacks. Pioneering work from Bell Laboratories [13] opened the possibility of a microtechnologies approach to the fuel cells area. Later, Jet Propulsion Laboratory (JPL) and Los Alamos National Laboratory (LANL) (in collaboration with Motorola) have created R&D activities targeted at increasing the mobile phone market.

JPL, in collaboration with Giner Inc., has demonstrated a miniature ‘‘flat-pack’’ DMFC stack capable of producing 150 mW continuously. The authors suggest a power requirement for cellular phones of 100–150 mW, during the stand-by mode and 800–1800 mW under operating conditions. The DMFC power source is positioned within the cellular phone, as in the case of the state-of-the-art lithium ion batteries, and the weight and volume of the device are about 50 g and 50 ml, respectively. The feeding of pure methanol is provided from a cartridge and is diluted prior to being fed to the anode; the reaction products are contained within the package. Air is delivered to the cathode by natural convection. As to the stack design, the cells are externally connected in series on the same membrane. Two "flat-packs", integrated in a back-to-back configuration, form a "twin-pack". Three "twin-packs" in series will be needed to obtain the power requirement of a cellular phone. The principal problems of this configuration are the ohmic internal resistance and the current distribution, this latter being rather non-uniform. The noble metal loading in the electrodes is 4–6 mg/cm^2. The principal goal of the JPL researchers is a 1 W DMFC power source, with the desired specifications for weight and volume and having an efficiency of 20% for a 10 h operating time, without replacement of methanol cartridges [14].

LANL has also been involved in the development of the portable DMFC power source. The first approach was to replace the "BA 5590" primary lithium battery, used by the USA Army in communication systems. The configuration is a bipolar five-cell DMFC stack, with electrodes having an active area of 45 cm^2 [15]. The small thickness (i.e. 2 mm) of each cell and a highly effective flow-field for air are the principal characteristics of this prototype. The main disadvantages are the high working temperature (60°C) and the low methanol concentration used (0.5 M) to reduce the cross-over rate. This implies difficult water management. The stack achieves 80 W peak power at a potential of 14 V, a power density of 300 W/l and an estimated specific energy of 200 Wh/kg assuming a weight of the auxiliaries twice the weight of the stack.

Another LANL activity, in co-operation with Motorola, is the development of small power DMFC for applications in cellular phones, laptop computers, portable cameras and electronic games [16]. The configuration selected for this prototype is planar, with four series-connected cells each with 5 cm × 5 cm area and 1 cm thick. The principal innovation of this concept is the multi-layer ceramic technology developed for the flow-field. The prototype exhibits an average power density of 15–22 mW/cm^2. Motorola's goal is the realisation of a fully automated miniature DMFC (100 mW power), with an energy density of five times that of the state-of-the-art lithium ion batteries.

A much-publicised development in this sector has recently been carried out by Energy Related Devices Inc., in alliance with Manhattan Scientific Inc. [17]. The main innovation introduced by this group is the utilisation of a semiconductor industry manufacturing approach, currently used for the production of microchips. A relatively low cost sputtering method is being used for the deposition of electrodes. The configuration appears to be planar with an external series connection having a thickness of about a millimeter. The use of semiconductor manufacturing technologies opens the way to different possible choices in geometry and arrangement of the whole device. The fuel distribution manifold is internal, whereas the air distribution manifold is external to the array. A wicking/diffusion mechanism is used to drive both feed as reactant and exhaust of product. A specific energy of 370 Wh/kg and an energy density of 250 Wh/l have been attained with this prototype.

Another interesting approaches has been made by the University of Minnesota [18] which is working on the realisation of miniaturised DMFC stacks on silicon substrates by using microelectronic fabrication techniques. Moreover, Stanford University [19] is examining the potentialities of a non-planar interface geometry, using photolithographic patterning, for the realisation of microfuel cells. Finally Fraunhofer Institute has already realised different prototypes in an integrated planar configuration [20].

At the present time, DMFC seems to be the most promising technology for applications in portable and micropower generation, due to the energy content and the liquid nature of the fuel. The introduction of semiconductor technology into the development of microfuel and minifuel cells likely will determine, in the near future, a revolution in the sector and a probable replacement of the most advanced type of rechargeable batteries with DMFC. The reasons for this are: (a) the high energy density of methanol; (b) the instant refuelling; (c) the challenge of further reduction of the weight and volume of the DMFC.

The on going improvements, by leading laboratories, in the development of unsupported catalysts [20], targeted for thin films MEA and the construction of the whole system architecture by using planar configuration with dc/ac converter to step up the potential to about 4–5 V will likely contribute to the achievement of targeted goals. Finally, in this particular market sector, the cost per kW or per kWh is already acceptable based on a scenario of mass production of micro fuel cells that could take advantage of established semiconductor technology for a progressive scale-up.

4. Fuel cell transport applications

In the last 10 years the major car manufacturers have begun launching intensive R&D programs on fuel cell vehicle (FCV). The reasons for this are that transport fuels must be better utilised in terms of efficiency and environmental polluting. The contribution of transportation represents one-third of global CO_2 emission to the atmosphere, so that important changes are expected from this sector in order to reach the aims of the Kyoto protocol.

FCs, obviously, match the requirements for vehicle applications since the absence of moving parts eliminate vibration and sound and the FCV do not suffer from the short range, weight, short battery life, and long recharge time suffered by batteries powered EVs. Also FCV can make use

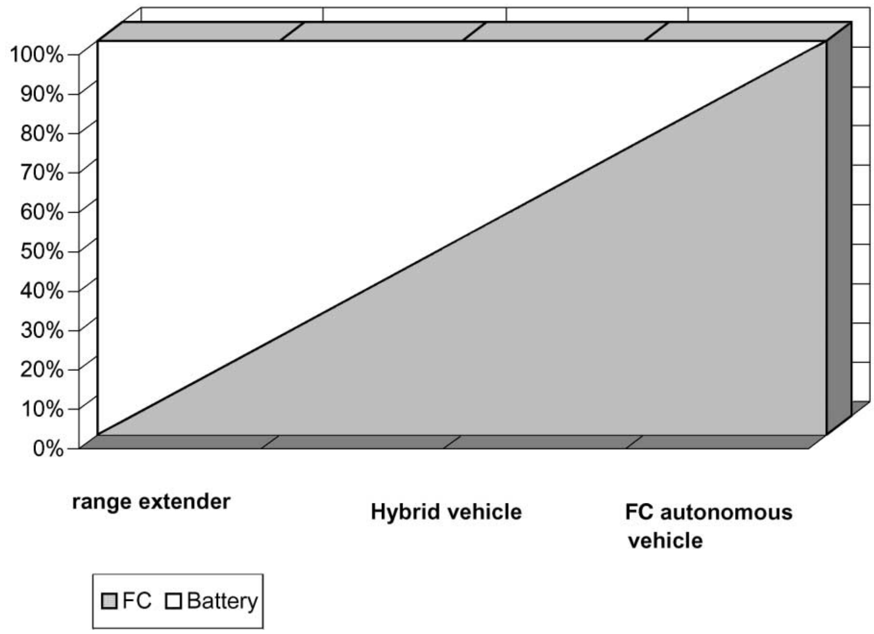

Fig. 2. Applications proposed for vehicles as a function of the degree of integration of the battery/fuel cell system.

of different fuels. However, a lot of problems still remain from the technical, economical and political view points and the next 4–5 years will be decisive in understanding the real role that FCV can play in the transportation sector.

A FC can be used in a car both as auxiliary power unit (APU) or as a power generator for an electric engine in a hybrid (FC + battery) or an autonomous configuration (only FC).

When the fuel cell works as an APU, the car uses an internal combustion engine (ICE) and part of the fuel is converted into electricity through the FC more efficiently than through the system using a traditional generator and battery.

Since the electricity needs for auxiliaries on-board (electronic devices, cooling system, control, etc.) are continuously increasing, the use of a FC as an efficient power generator is very important, but the most important contribution that this technology can make is as a power generator for an electric engine. In this case, the different degree of hybridisation depends on the type of vehicle and on its mission, but depends also on the technical and economical development of the fuel cell system.

As shown in Fig. 2, a FCV is realised when the system is totally driven by a FC with a very small contribution from a battery that is used during the start-up or for power peak and to recover the braking energy; when the FC power is less than the 25% of the engine power, the range extender configuration is realised in which the FC is used to recharge the battery [21].

Two main types of FCV have been proposed: the direct fuel cell vehicle (DFCV) in which the fuel is electrochemically oxidised inside the FC and the processed fuel cell vehicle (PFCV) where the fuel is first converted into hydrogen by a fuel processor and then the hydrogen is fed to the FC to produce electricity (see Fig. 3) [22].

4.1. Direct fuel cell vehicle

In the case of the DFCV, the FC technology is expected to be PEFC if fed by H_2, or DMFC if methanol is to be the fuel.

In both cases, CO_2 emissions are considerably lower than for a vehicle using a gasoline ICE and the efficiency is greatly improved. As shown in Fig. 4, the DFCV fed with hydrogen represents the best solution since, in fact, the efficiency is more than double and the CO_2 emissions lowered by two-thirds with respect to an ICE if the emissions for hydrogen production from the fossil fuel is considered. Of course the CO_2 emission is zero when hydrogen is obtained from renewable sources.

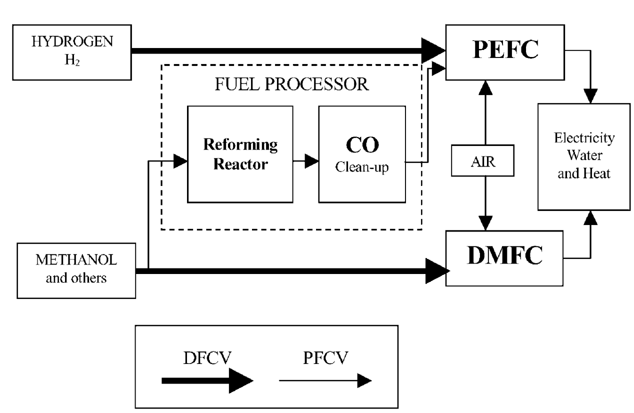

Fig. 3. Functional scheme of direct (DFCV) and processed (PFCV) fuel cell vehicle systems.

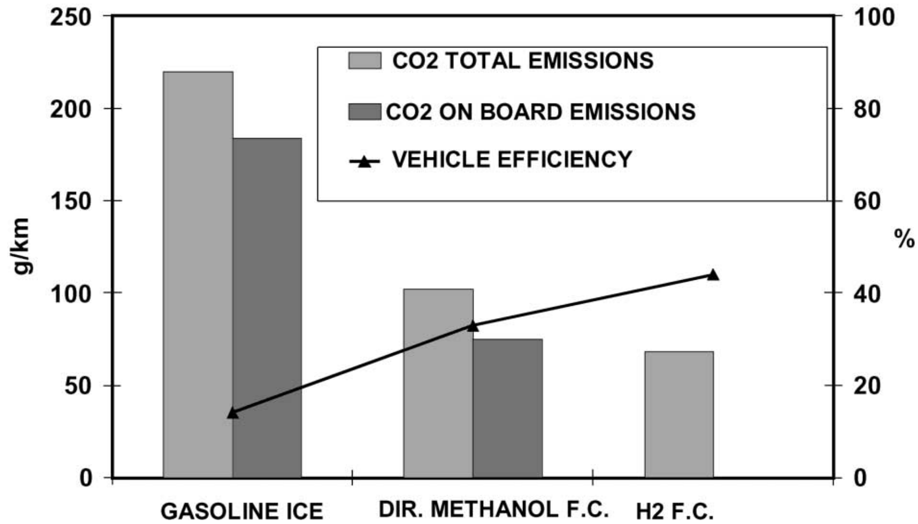

Fig. 4. CO_2 emissions and efficiency for traditional ICE and methanol or hydrogen fed fuel cells.

Even if the emissions and the efficiency of a DFCV perfectly fit with the sector demand, many problems have to be solved before commercialisation. Among these, special attention is devoted to fuel production and infrastructures. In fact, when a new fuel is proposed, the infrastructure must serve only a few vehicles at the beginning but thousands or millions once the technology reaches maturity. There is great difficulty in bringing a new infrastructure and new vehicles to the market place at the same time. From this point of view, hydrogen is worse than methanol. Costly changes are needed to make methanol widely available at filling stations, but for hydrogen even expensive changes are foreseen. For this fuel, the evolution of this fuel will be slow initially hydrogen might be produced and stored at the filling stations by means of small on-site processors, based on steam reforming or on partial oxidation of natural gas. Obviously hydrogen could be produced also from methanol, ethanol or gasoline, and then it could become one of the possible fuels available at future stations. When hydrogen vehicles become more widely used, larger transformation stations, also using electrolysers, could be developed and the hydrogen could be distributed by pipeline or by liquid tankers. In this possible scenario, the weak point is the cost-effectiveness of small hydrogen generators, while social benefits, coming from lower emissions and higher efficiency, added to the fuel flexibility that this prospect introduces, giving the opportunity for exploitation of local resources, represent the strong points [23].

Turning to methanol, there is currently no central distribution infrastructure for this fuel but results of different studies indicate that: "...the existing gasoline retail distribution system can be adapted to serve methanol with few modification..." [24], and "...methanol station can cost between US$ 17,000 and 70,000 per station and the equipment is very similar to a gasoline system with special attention to ensure methanol compatible components are used" [25]. This means that if 10% of fuel stations were to be converted to methanol in Europe, the total cost would be US$ 590,000,000.

Methanol is generally obtained from natural gas and in the future it can be produced starting from biomass; as it is liquid at atmospheric pressure and temperature, it can be easily transported by track, rail car, ship and also by pipeline. As the methanol "supporter" says, it will be handled in much the same way as gasoline.

Assuming that governments and international protocols will play their part in accelerating the elimination of infrastructure obstacles, one technical problem remains for each type of DFCV under consideration for vehicles and can influence the time for commercialisation: on-board hydrogen storage and the DMFC stack.

The on-board hydrogen storage system is the main technological obstacle for the development of direct hydrogen fuel cell vehicles. To overcome this difficulty, different technical solutions have been proposed, some ready for the commercialisation, some still at the research and development stage but with good energy density values that are comparable to today's liquid fuels.

In current zero-emission vehicle prototypes, containers of novel composite materials are used to store gaseous hydrogen at 200 atm with a capacity storage of 0.5–2 kWh/kg. This system is based on simple and cheap technology and is now well established. Current containers consist of carbon/glass/aramidic fibres that weigh three or four times less than metallic containers, and their strength offers high safety potential in case of crash. Nevertheless, they need a large volume and containers still represent low specific energy storage. Therefore, this method is only suited for larger vehicles like buses or vans. Cryogenic hydrogen has many advantages over pressurised hydrogen, providing an energy density of 6 kWh/kg. Drawbacks of liquid hydrogen are a greater system complexity on-board and for refuelling.

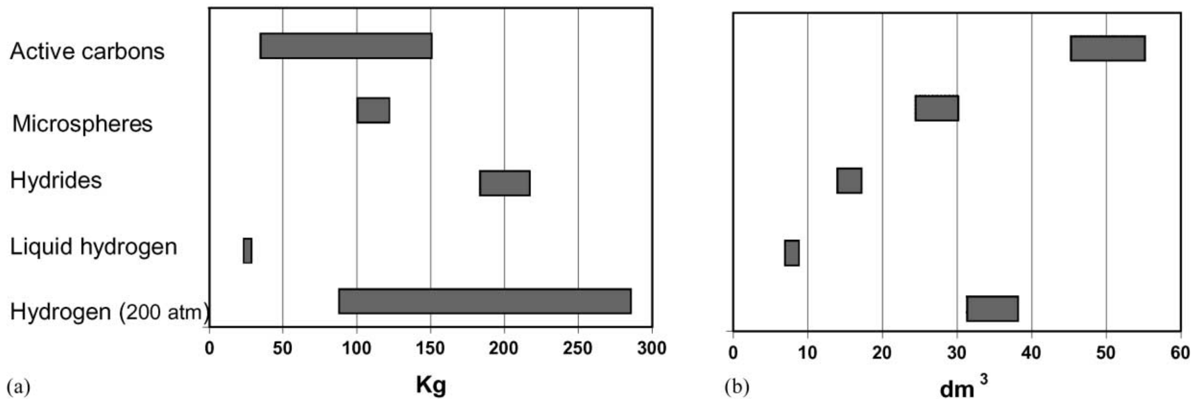

Fig. 5. Weight (a) and volume (b) needed to store 4 kg of H_2 (400 km of estimated autonomy) for different means.

Cooling of hydrogen to achieve liquefaction consumes about one-third of its energy content, while the energy consumption for pressurised hydrogen is only 4–7%. Therefore, cryogenic hydrogen is only suited for concept cars and prototypes.

Hydrogen can be bonded chemically to metal atoms to form metal hydrides that offer lower pressures and higher energy density than gaseous hydrogen, and comparable with the liquid state. The main drawback is the weight: the total amount of hydrogen absorbed is generally 1–2% of the total weight of the tank. Some metal hydrides are capable of storing 5–7% of their own weight, but only when heated to temperatures of 250°C or higher. Therefore, for the same weight this system has a capacity three/four times lower than pressurised hydrogen. Because of its low pressure and high stability, the hydride storage system can be packaged into any size and shape which allows hydrogen to be transported safely and easily.

Tiny hollow glass spheres can also be used to store hydrogen safely. The glass spheres are warmed, increasing the permeability of their walls, and filled by being immersed in high-pressure hydrogen gas. The spheres are then cooled, locking the hydrogen inside the glass balls. A subsequent increase in temperature will release the hydrogen trapped in the spheres. Microspheres have the potential to be very safe, to resist contamination, and to contain hydrogen at a low pressure, increasing the margin of safety. Their specific energy is around 1.2 kWh/kg.

Low pressure systems for stored gas adsorbed on activated carbon are much more attractive than high pressure systems. The low pressure storage system can be packaged into any shape which allows hydrogen to be transported easily in small tanks. The system needs a refrigerating system, therefore it is more complex than cryogenic hydrogen but has a higher energy density (1 kWh/kg) and it is cheaper.

In recent years, carbon nanotubes have attracted interest due to reports of enormous hydrogen storage capacity. Carbon nanotubes are microscopic tubes of carbon (2 nm diameter) that store hydrogen in microscopic pores on the tubes and within the tube structures. Similar to metal hydrides in their mechanism for storing and releasing hydrogen, carbon nanotubes after the advantage of being able to store a lot of hydrogen. Data published in the past claimed sensational capacities of up to 60 wt.% [26]. Unfortunately, these results were not reproducible. Now, several research groups expect the storage capacity to be in the range of 20–40 wt.% [27,28]. It is not yet clear which modification of nanotubes has the best storage capacity. Further studies are needed to show whether carbon nanotubes are a serious alternative to other storage forms. They are still in a research and development stage. Research on this promising technology has focused on the improvement of manufacturing techniques and cost reduction as carbon nanotubes move towards commercialisation.

Fig. 5, shows a practical comparison of the weight (a) and volume (b) needed to store 4 kg of hydrogen (which is the amount needed for a DFCV with a 400 km of autonomy), by means of the different storage technologies, which are listed on the *y*-axis in order of their distance from commercialisation. Finally, in Fig. 6, several hydrogen storage technologies are compared, in terms of weight percentage of stored hydrogen. From these figures, the necessity for more research and developing activity in this sector is evident.

The FCV could also make use of methanol as fuel, and here the main technological problems are related to the FC

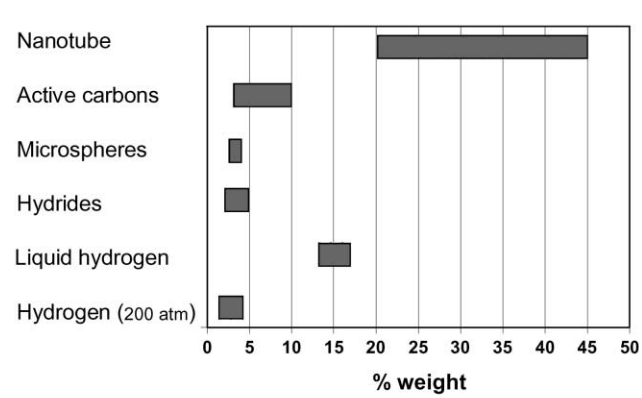

Fig. 6. Percentage by weight of stored hydrogen for different means.

Table 4
Some recent results obtained from research groups working on DMFC

Stack developer	Application[a]/ year	Rated power (W)	Maximum power density (mW/cm^2)	Specific power density (W/kg)	Specific power density (W/l)	T (°C)	Oxidant/P (atm)	Anode catalyst and loading (mg/cm^2)	Cathode catalyst and loading (mg/cm^2)	No. of cells/ surface area (cm^2)
Siemens Johnson–Matthey IRD	T/2000	850	100	–	–	104	Air/1.5	Pt-Ru/C/1.3	Pt black/4	16/550
Los Alamos	P/2000	17	75	–	300[b]	60	Air/0.7	–	–	5/45
Los Alamos	T/2000	47	220	–	1200[b]	100	Air/3	Pt-Ru black/1.2	–	5/45
KIER	T/1998	40	90	–	–	90	Oxygen/3	Pt-Ru/C/2	Pt/C/3	3/150
Sodeteg–Nuvera–CNR–TAE Thomson LCR	T/2000	150	150	150[c]	210[c]	110	Air/3	Pt-Ru/C/2	Pt/C/2	5/225

[a] T: transportation; P: portable.
[b] Calculated with respect to the active electrode surface.
[c] Calculated on the basis of the overall stack device.

stack. In fact, as is well known, a DMFC is essentially the same as a PEFC, but it is supplied directly with methanol which strongly slows the kinetics of the fuel electro-oxidation, with consequent lower performances compared to PEFC. To increase the performance, a higher catalyst loading is used, with higher costs in comparison to PEFC, and the system is forced to operate [29] at higher temperature (100–130°C).

A severe problem for DMFC is the fuel crossover through the membrane. This effect, caused by the permeability of the membrane to the aqueous fuel mixture, lowers the FC efficiency markedly; however crossover can be reduced with suitable membranes when a higher electric load is used. The latest improvements in developing new membranes for DMFC have reduced the crossover from 50 to about 10%.

Some recent results obtained from research groups working on DMFC are reported in Table 4. Although the results are obtained under different conditions it is evident that higher power density can be obtained at high temperature due to the improved methanol electro-oxidation kinetics. Since the state-of-the-art membrane dehydrates at temperatures above 100°C, new membranes, capable of sustaining high temperature (150–160°C), with ambient humidification and with low methanol crossover, must be developed [30].

It will also be necessary, to develop new components and stack technology in order to reach the technical targets reported in Table 5. These activities will have to include the reduction in the noble metal content of the catalyst, the development of thin film electrode structures and the optimisation of the whole MEA.

Table 5
The 2004 technical target for new components and stack technology for transportation applications

Target for 2004	Integrated system PEFC + processor	DMFC
Efficiency (%)	48	50–60
Power density (W/l)	300	300
Specific power (W/kg)	300	300
Costs (US$/kW)	50	45
Endurance (h)	5000	5000

4.2. Processed fuel cell vehicle

While waiting for the DFCV, an interim solution claimed to enter the market, within 2004–2005, is the so-called processed fuel cell vehicle (PFCV), a vehicle system with FC stacks fed with H_2-rich gas produced on-board by a fuel processor (FP) supplied by a primary fuel transported inside the vehicle. The composition of the gas produced by the FP is an essential issue for efficient operation, as the presence of CO causes a rapid decrease of FC performance. An on-board FP usually lowers the overall efficiency of the system and requires high FC catalyst loading, resulting in higher costs and increases in system weight and volume. Nevertheless, it allows the use of different fuels (methanol, propane or gasoline) in a multi-fuel configuration.

For the most recent PEFC stacks the power density is in the range of 0.8–1 kW/kg and 1.2–1.5 kW/l [31]. As reported above for DMFC, the higher the temperature the faster are the reaction kinetics and higher the specific power; thus one of the aim of PEFC stack [31] commercialisation involves the development of new membranes able to work at temperatures over 90–100°C. The operating pressure is also a debated question; in fact, high pressures improve the gas diffusion characteristics of the MEA and consequently the specific power but, on the other hand, noisy, heavy and inefficient air compressors are needed [32].

The integration of a FP in the FC stack of a PFCV remains the main problem, because the FP, operating on-board, needs to satisfy the following several requirements: (i) start-up time less than a few minutes; (ii) load transient response less than 500 ms; (iii) fast feeding of the FC stack [33].

Steam reforming, almost has been abandoned and the candidate processes for on-board FP are partial oxidation and autothermal reforming, which is a combination of exothermic partial oxidation and endothermic steam reforming.

In both cases a further purification phase is needed for a complete CO clean-up.

The system efficiency, in terms of fuel conversion to hydrogen, CO production and the general management of the interconnected system of the energy-fed electric engine, depends on the fuel. Methanol has been the first fuel to be considered due to the wide experience gained from stationary applications. The development of a gasoline FP is the goal of several companies, and probably the future of the fuel strategy will depend on the results of this activity.

Obviously the gasoline for FP will be formulated differently with respect to the fuel currently available but the possibility of using gasoline in FCs will imply smaller and cheaper infrastructure modifications with respect to those that would be needed for methanol.

Thus, the interim solution is not completely clear; methanol or gasoline?

Obviously, this does not depend only on technical factors: the huge automobile market, the large interest in fossil fuel production and the capability to be ready for a prompt or slow change toward a hydrogen market will all condition the final choice. In the meantime many prototypes have been realised and alliances among car-makers, fuel producers, fuel distributors, and fuel cell producers have been formed both to verify the new technology on the road and to indicate the way forward.

4.3. State-of-the-art on transport applications and remarks

Research programs such as the PNVG (partnership for a New Generation Vehicle) in the USA, the California Fuel Cell Partnership, Hydro-Gen, Capri, FCBUS in Europe are promoting FCV demonstration activities, but the alliances, mentioned above, play an important role in the rational development of prototypes. The most important and well known alliance is that between Ballard, Daimler–Benz and Ford, established in 1997, with the aim of developing the fuel cell engine in competition with other producers such as the Toyota, Honda and GM/Opel groups. From co-operative research projects or specific agreements between automakers and fuel producers or distributors, two strategies are emerging: the first, guided by Daimler–Chrysler, encourages the use of methanol as the fuel for cars, but does pay attention to the development, with Shell Hydrogen, of gasoline reforming so that a multi-fuel PFCV might be realised in the future; the long term goal is the use of methanol and hydrogen. The second strategy, guided by GM/Opel, allied with Exxon, is mainly concentrating on gasoline, avoiding any new fuel, which might be refused by customers, and pose for infrastructure problems, and in this case the long term goal is hydrogen alone.

The first group able to commercialise a FCV will gain a great advantage over competitors, but at the same time the risk margins remain very high.

Daimler has produced two models called Necar4 and Necar5; the first is a sample of DFCV fed with liquid hydrogen, while the second is a PFCV with reformed methanol. This company plans to commercialise FCV in 2004 in line with the goals of Toyota, Mazda, Honda and Ford.

In the meantime GM/Opel has recently presented the Hydrogen1, a DFCV fed with liquid hydrogen, with good performance.

Special mention should be made of the Fuel Cell Bus, which has the advantage of representing an important specific market, capable of introducing the concept of hydrogen as an energy vector for transportation. In this sector two important activities have been developed, in Chicago and Vancouver, where two fleets have been tested for more than 100,000 km. Numerous prototypes have been built, mainly based on PEFC stacks fed with hydrogen stored under high pressure. This type of application is important because it fits the possibility of on-site hydrogen production (i.e. from methane) and re-filling in the same location.

There are huge financial resources available for FCV development; this will likely result in a decisive push to the introduction of fuel cell technology.

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