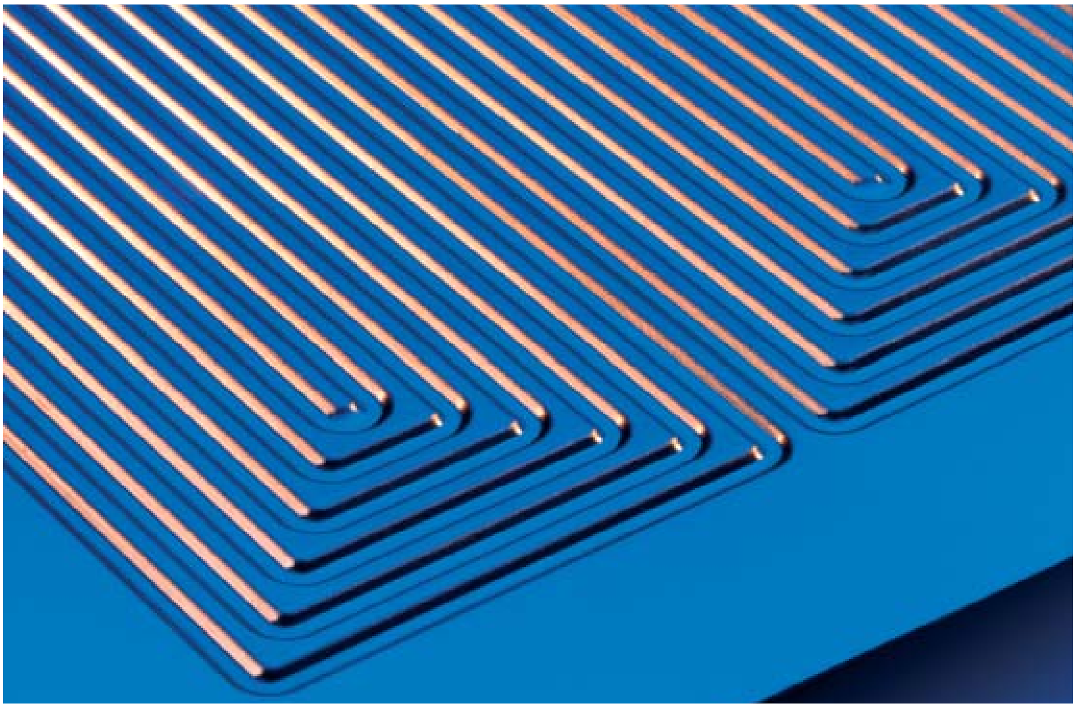
Innovative concepts for bipolar plates

Industrial development of fuel cells of the PEMFC type requires major advances to be achieved with respect to cost, for their various components. In particular, bipolar plates still involve a prohibitive cost, precluding widespread utilization. Drawing on its expertise in the areas of composite and metallic material formulation and fabrication, CEA is developing two novel plate concepts: stamped sheet metal, and molded organic composites. Further investigations at CEA concern gas distribution by means of metal foams, a path that appears promising.



Bringing the cost of fuel cells down entails development of new bipolar plate concepts. The picture shows a stamped-metal bipolar plate.

D. Michon-Artechnique/CEA

A global approach of PEM fuel-cell systems

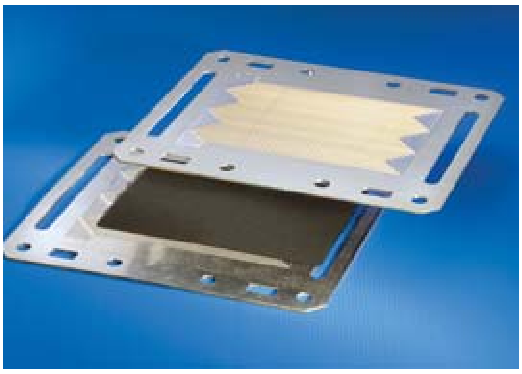
For the development of fuel-cell systems, CEA implements a four-stage, global approach, starting with functional analysis and **modeling** of the system comprising the plates and related components. The second stage consists in identification and investigation of physical parameters, elementary processes and their interactions, liable to impact operation, performance and optimized sizing of components in the basic cell, and in the system. The next stage involves the design (architecture and sizing), construction, bringing into operation, and evaluation of demonstrators (scale models, elementary components, and full-scale modules) and related fabrication processes. As and when required, investigations are carried out, of materials (selection, **formulation**, characterization) and processes (fabrication, forming, assembly, coatings, surface treatments).

The aim? Seeking to define, and validate, resources enabling fast, comparative evaluation of materials, surface states and coatings by means of *ex-situ* tests, or tests in relevant simulated environments, on the basis of a thoroughgoing analysis of in-cell trials in actual conditions. This rigorous, proactive approach is not exempt from surprises and mishaps, which do bring about, in the longer term, a significant enhancement in knowledge and understanding of related domains, and of PEM fuel cells in general.

The bipolar plate should be considered at one and the same time in its threefold aspect. First of all, it forms the interface between the system and the core of the **basic electrochemical cell**. Second, it is an integral part of the basic repeated structural unit – together with seals and MEAs – and, as such, strongly impacts both sides of the cell's cost/performance ratio. Finally, it is the decisive component as regards cell or module characteristics, in terms of architecture, geometry, volume, mass, and mechanical characteristics.

Bipolar plates serve as gas distributors (**hydrogen**, oxygen or air) and current collectors. Their materials must meet demanding specifications, with respect to **electric** and **thermal conductivity**, impermeability to gases, mechanical properties (resistance to tightening stresses), and chemical and thermal properties, in the prevailing operating conditions for cells of PEMFC or DMFC type.

Use of **graphite** plates meets these criteria. However, the requirement to machine channels into the plate's surface, for gas transport and removal of water forming inside the cell, results in further cost, incompatible with volume commercialization of power cells (average pricing of €1/cm^2 for a machined graphite



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Stamped-metal bipolar plates and membrane–electrode assembly. The MEA sandwiched between two plates constitutes a single cell. Surface grooving on the plates allows supply of hydrogen on one side, and oxygen (or air) on the other side. A cooling fluid circulates between the two back-to-back, stamped-metal distributors.

plate, with variations depending on channel geometry and fluid inlet/outlet type).
Two novel low-cost bipolar plate concepts are consequently being developed at CEA.

Stamped sheet metal

Work carried out at CEA, over the past few years, on bipolar plates, with regard to cost reduction, improved performance, reliability, and durability, has already enabled cuts in mass and volume by 3, and the bringing down, by a factor 100, of prospective pricing for this component, compared with conventional machined graphite plate technology. (1)
To achieve this, research workers at CEA/Grenoble implemented, for metal distributors or bipolar plates to be used in PEM fuel cells, an approach that is at the core of their overall stance, as regards development of fuel cells and their components (see Box). This approach led them to devise innovative architectures, and develop and use appropriate testing apparatus, and rigorous test and evaluation protocols, together with the required test rigs. Among the innovative architectures involved may be noted plates featuring serpentine channels and integral liquid cooling system, based on back-to-back assembly of stamped metal sheets (see Figure 1), one-piece, metal-foam distributors… The test rigs constructed bring together stamped-metal single-cell units of 60 cm² surface area, in a variety of stainless steels; a reference cell, using graphite machined to the same geometry; full-scale, 120-cm², 240-cm² and 360-cm² useful area, 3-, 6- and 9-channel stamped-metal plates, cooled by internal fluid circulation; and 1–2 kW demonstration modules. Test and evaluation protocols include static and cyclic endurance tests, and dynamic tests (see Figure 2).
In this manner, taking on board the constraints specific to stamping led to successful exploration of channel-profile geometries that had been ignored by conventional machined graphite plate technology. The carrying through of rigorous comparative tests of various standard alloys, in stamped sheet form, in a

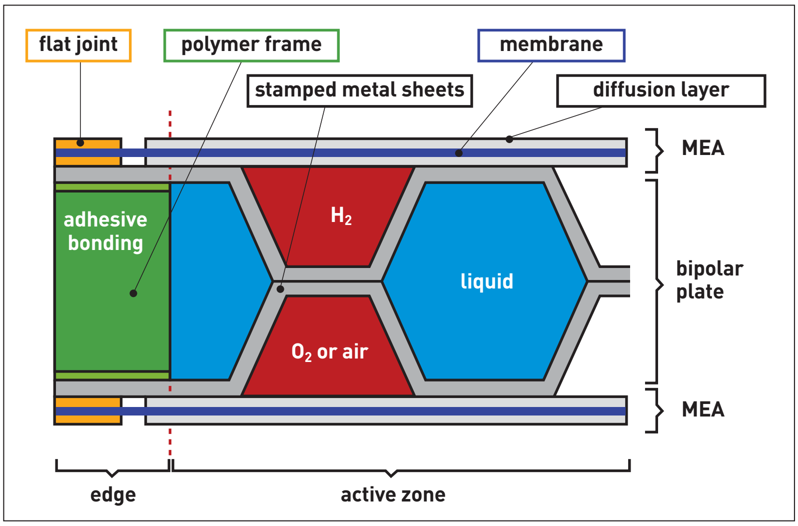
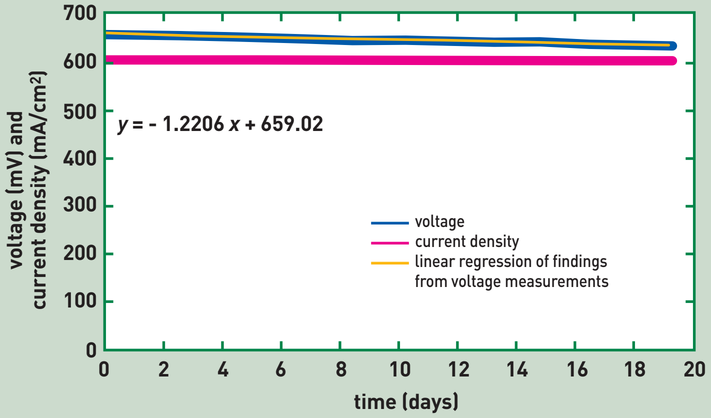
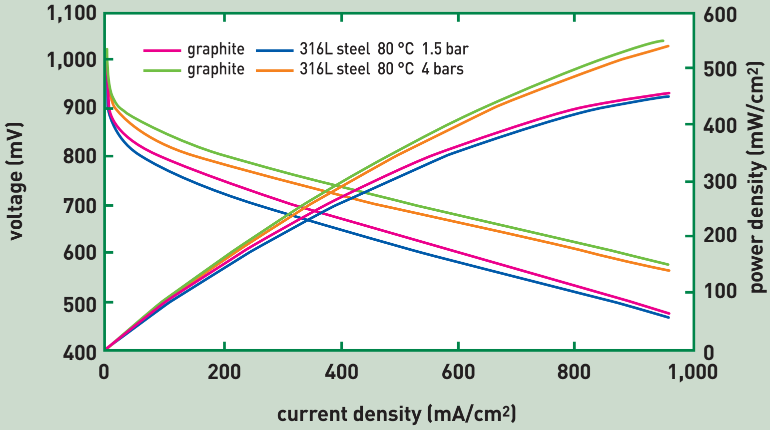


Figure 1.
Concept for a cooled, stamped-metal bipolar plate architecture.



test conditions: 80 °C, 4 bars, H_2/O_2 stoichiometry 1.2/1.5, humidification T_{sat} = 85/90 °C, i = 600 mA/cm², MEA: Nafion ® 115 + Etek electrodes assembled by Nafion ® deposition + hot pressing.



test conditions: H_2 stoichiometry = 1.2, O_2 stoichiometry = 1.5, H_2O_2 100% moisture content, curve scanning rate 0.017 A/s, $i_{\min O_2}$ = 125 mA/cm², $i_{\min H_2}$ = 125 mA/cm².

Figure 2.
Findings from tests on a single-cell unit. Top, static endurance test on a 60-cm², 2-parallel-channel single cell using stamped 316L stainless steel. Bottom, intensity/voltage curves for a single cell using stamped 316L stainless steel, and a reference cell using graphite machined to the same geometry, for pressures of 1.5 bar and 4.0 bars (dynamic test).

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(1) Part of the work described here was carried out as part of PREDIT–Ademe contract No. 99 74 003 (PREDIT: Programme de recherche et d'innovation dans les transports terrestres – French government Research and Innovation Program for Land Transportation).


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Automated simultaneous test bench for 4 independent single cells. Top, temperature-regulated test cells. Bottom, general view.

single cell – long thwarted though this had been by differences in stamping behavior between materials – allowed the key influence to be appreciated, of channel depth, related to the module's feed and operating parameters, and an understanding to be gained of the properties of the diffusion layers, as well as of MEA performance dispersion. Analysis of the causes of dispersion in the findings further led research workers to a thoroughgoing reassessment of module tightening conditions and technologies, greater awareness of the widely underestimated importance of the seals, and identification of a hitherto-unknown mechanism involved in cell water management, greatly impacting cell performance in some conditions. Initial in-cell comparative endurance tests, in cyclic conditions, of stamped-metal and machined-graphite distributors revealed a swift degradation, in cyclic conditions, of MEAs selected on the basis of short-term performance! In-cell static endurance tests showed a very moderate degradation for fuel cells using 316L stainless steel, contributing one third of MEA degradation (see Figure 2).

This small group of findings, arrived at in the context of the global approach implemented at CEA, evidence a potential for fruitful interaction with approaches favored by manufacturers, generally more focused, owing to specific constraints, and pursuit of shorter-term objectives.

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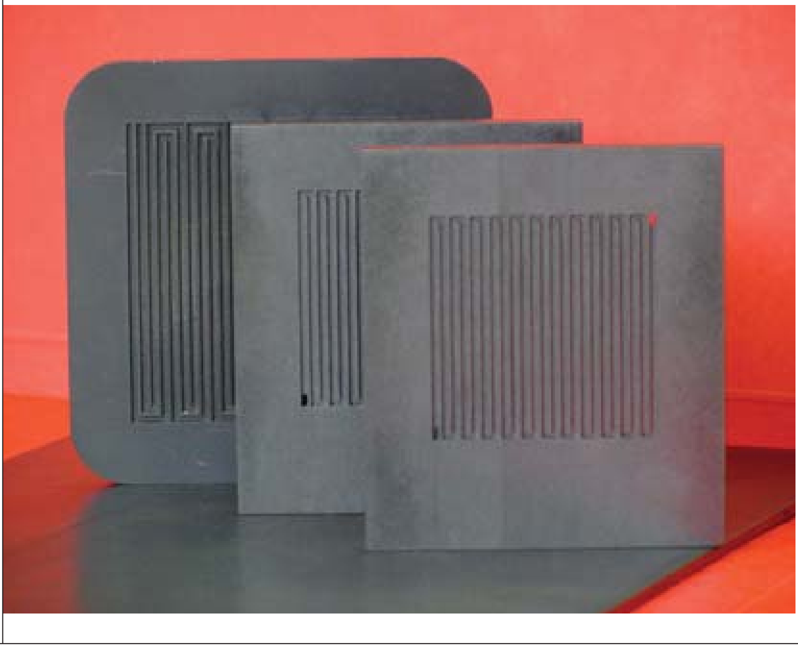
Molded organic composites

The solution here consists in dispersing into a **thermoplastic** or **thermosetting** matrix conducting, noncorroding fillers, such as graphite platelets, carbon fibers or carbon black. At high volumetric contents, these electrically conducting constituents confer onto the **composite** thus formed **electric** and **thermal conductivity** properties close to those of graphite plates.

Further, such composite **formulations** allow direct molding of bipolar plates along with their channel system, or even with the fluid inlets and outlets. The gain accruing from this technology represents cost savings by a factor 10–100, compared to the machined-graphite plate path.

Molded composite bipolar plates.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Though this solution has been the target of major research and development efforts from manufacturers and research organizations, (2) fabrication conditions for these composite materials remain demanding, in terms of molding time, and hence costly.

CEA (Le Ripault Center) and Atofina (Total Group) have initiated a joint effort, to develop an innovative microcomposite composition, a polyvinylidene fluoride- (PVDF) and graphite platelet-based material, optimized in terms of the physical properties required for bipolar plates. The original feature lies in the dispersion method for the PVDF and graphite particulates, yielding a homogeneous molding powder, readily transformed when heated.

With the powder molded by thermocompression, the physical properties of the composite plates obtained are highly attractive: high electric conductivities, very high thermal conductivities, and low permeability to helium (see Table).

physical properties	values
electric conductivity (S/cm)	250–350 in plane 30 through material
thermal conductivity (W/m/K)	110 in plane 12 through material
permeability to helium ($m^3 \cdot m/m^2/Pa/s$)	$3.7 \cdot 10^{-17}$ (measured at 80 °C and 2 bars absolute)
ultimate bending stress (MPa)	35

Table.
Main characteristics of composite plates obtained from thermocompression molding.

The specific geometry and structure of the microcomposite particulates make for easy composite forming by **extrusion**, this being true up to graphite contents higher than 80% by volume. This property thus makes it possible to consider fabrication by way of extrusion–compression. This molding technology, combining a continuous step – extrusion – and subsequent brief compression step, is found to be faster and more flexible than rival solutions. It should enable overall plate fabrication costs of around €20–30/kW to be achieved, i.e. about €0.01/cm^2, while preserving performance levels equal to those of materials fabricated by thermocompression.

This bipolar plate concept is subject to three patent applications, one of which is joint CEA–Atofina intellectual property. (3) Ongoing investigations are addressing development of the extrusion–compression technology, broadening the range of plate geometries and assemblies amenable to direct molding, and integration of seals and cooling, to produce complete components, ready for operational use.

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(2) These include SGL Carbon, Schunk, Morgan Fuel Cell, du Pont de Nemours, the Fraunhofer Institute, the Gas Technology Institute…

(3) "Poudre micro-composite à base de plaquettes de graphite et d'un fluoropolymère et objets fabriqués avec cette poudre" ("Graphite platelet- and fluoropolymer-based microcomposite powder, and objects manufactured from this powder"), joint CEA–Atofina intellectual property, application PCT/FR02/04569.

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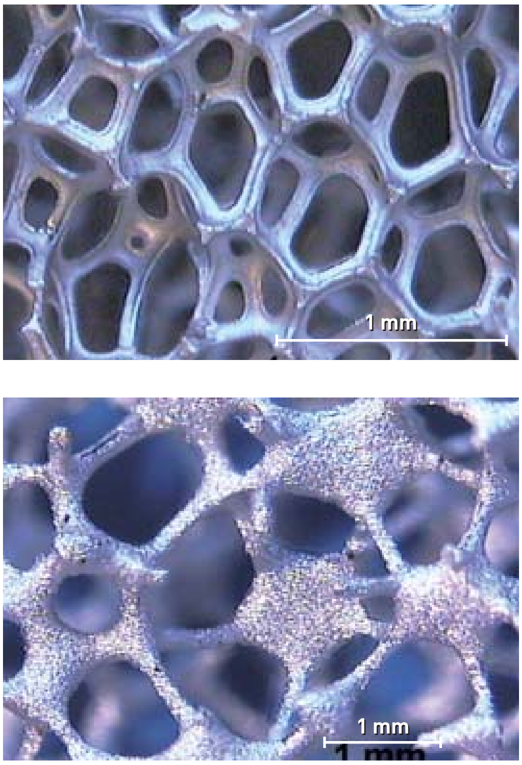
Extrusion–compression machine for fast molding of thermoplastic composites.

A promising concept: porous materials

Distribution of gaseous reactants and removal of the water formed are the key aspects of PEM fuel cell operation. Use of open-pore metal foams, to substitute conventional channel-carrying structures, is a promising alternative. Indeed, the high porosity of such materials favors gas accessing the **electrodes** (accessible surface area greater than 90%, rather than 60–70% for channels), allowing, it may be surmised, optimum utilization of **catalyst**-bearing surfaces. For the same reason, these materials may contribute to weight savings for the assemblies. Their use may further make for easier integration of some functions, such as cooling.



CEA – (NITECH product)

CEA – (PORVAIR product)

High-porosity metal foams are an attractive alternative for the distribution of gaseous reactants and removal of the water generated in PEM fuel cells.
Top, nickel foam;
bottom, stainless steel foam.

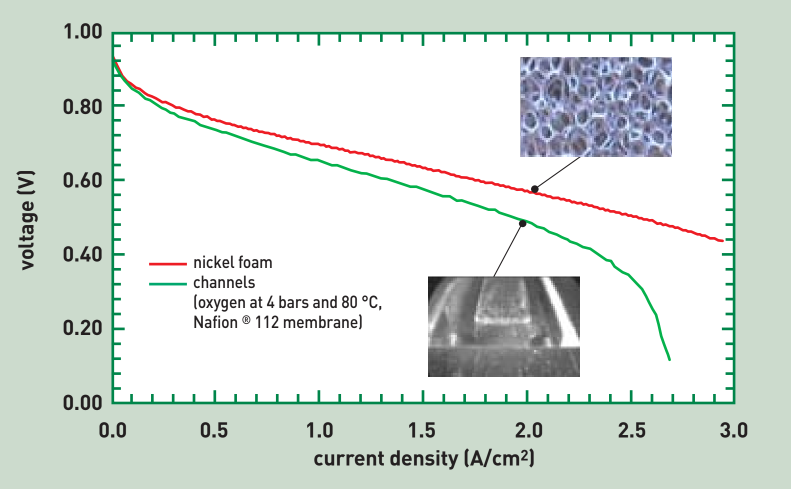


Figure 3.
Performance of a PEM fuel cell operating with various types of distributor, as indicated by output voltage as a function of **current density**. Nickel foam yields the best performance.

Exploratory tests on single cells have confirmed the advantages of metal foams, which, in similar conditions, allowed, over several tens of hours' operation, better performance to be achieved than with channel distributors (see Figure 3). These initial tests were carried out with 2-mm thick nickel foam, featuring pores of about 500 **µm**. However, with such a material, degradation of electrical response is found to set in over time, this process even, in some cases, accelerating over shutdown periods. This deterioration is caused by corrosion of the solid matrix, which may in particular induce contamination of the **electrolyte** membrane by metal **ions** from the porous material. Thus, in-cell behavior of metal foams is the critical point in this technology. The choice of material to form the foam is of prime importance, however this is often bound up with the fabrication process (**electrolysis**, deposition by **PVD**, **CVD**...). When the material used is susceptible to corrosion in a fuel-cell environment, one cure consists in overlaying it with a coating, of chromium for instance. A number of coatings have been tested on nickel foams, yielding sufficiently encouraging results to warrant considering construction of a realistically-sized trial stack (3 x 225-cm² cells). Nevertheless, identification of the corrosion mechanisms involved appears essential, as guidance for the choice of new materials, or even for the definition of shutdown protocols for systems using porous metallic materials.

Concurrently, in order to arrive at rules for distributor sizing (pore size, thickness, fluid inlet and outlet configuration), an investigation of flows in porous media of the foam type has been initiated. This relies on physical and numerical **modeling**, supported by trials on hydraulic mockups. These investigations have made it possible to put forward a model that may apply to such porous media, in the case of gaseous flows – consideration of **two-phase** (water–gas) aspects being addressed at a subsequent stage. Indeed, as in channel structures, presence of water generated by the cell is liable to impede access of reactants to the reaction sites. Further, owing to the small dimensions involved, **capillary** processes take on added importance in porous materials, increasing risks of water retention.

CEPACPOR stack, comprising three 225-cm^2 cells with metal-foam distributors, fitted with current-density measuring probes.

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Distribution of reactants by means of metal foams remains a promising path, in particular as regards increasing **power densities**. It does however require better understanding in the fields of corrosion and two-phase hydraulics, if it is to offer a viable alternative to channel distribution.

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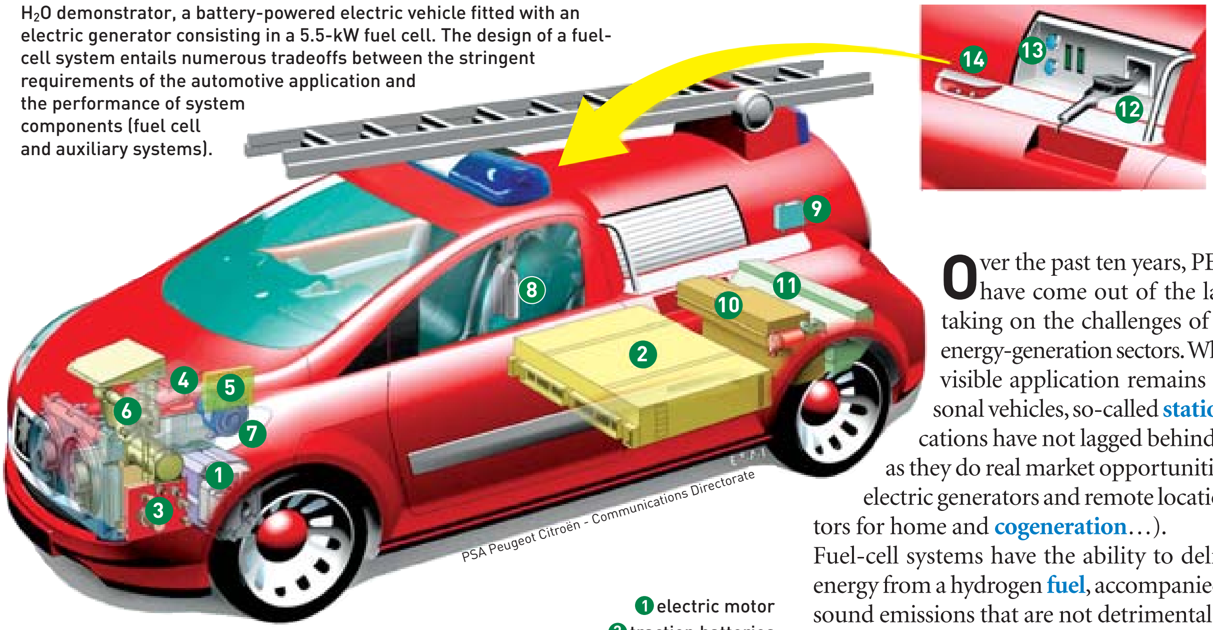
Fuel-cell system design: application to the automotive case

Concurrently with research work and technological innovations concerning improvements to the various components going into PEM fuel cells, investigations are ongoing on optimizing integration of these fuel cells into energetic overall systems. Definition of a fuel-cell system is the outcome of an ensemble of tradeoffs between requirements of the target application, of which the automotive application is the exemplar, and the performance of the various system components.



H_2O demonstrator, a battery-powered electric vehicle fitted with an electric generator consisting in a 5.5-kW fuel cell. The design of a fuel-cell system entails numerous tradeoffs between the stringent requirements of the automotive application and the performance of system components (fuel cell and auxiliary systems).

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① electric motor
② traction batteries
③ fuel cell (complementary power source)
④ electric vehicle control computer ⑤ hydrogen-generator computer
⑥ hydrogen generator ⑦ air pump ⑧ compressed-oxygen flasks
⑨ safety management computer ⑩ fuel tank (sodium borohydride)
⑪ recycling tank (borax) ⑫ outside power uptake
⑬ electric connections for auxiliary systems ⑭ refueling panel.

Over the past ten years, PEM fuel cells have come out of the laboratories, taking on the challenges of a variety of energy-generation sectors. While the most visible application remains that of personal vehicles, so-called **stationary** applications have not lagged behind, presenting as they do real market opportunities (portable electric generators and remote locations, generators for home and **cogeneration**…).
Fuel-cell systems have the ability to deliver electric energy from a hydrogen **fuel**, accompanied by gas and sound emissions that are not detrimental to the environment. Some of the performance requirements for such systems are the same, whichever application is targeted: high energy **efficiency** (> 40%), good durability (> 5,000 hours for the automotive case, and indeed 10,000-40,000 hours for stationary applications), low pollutant emissions, and reasonable outlay costs. On

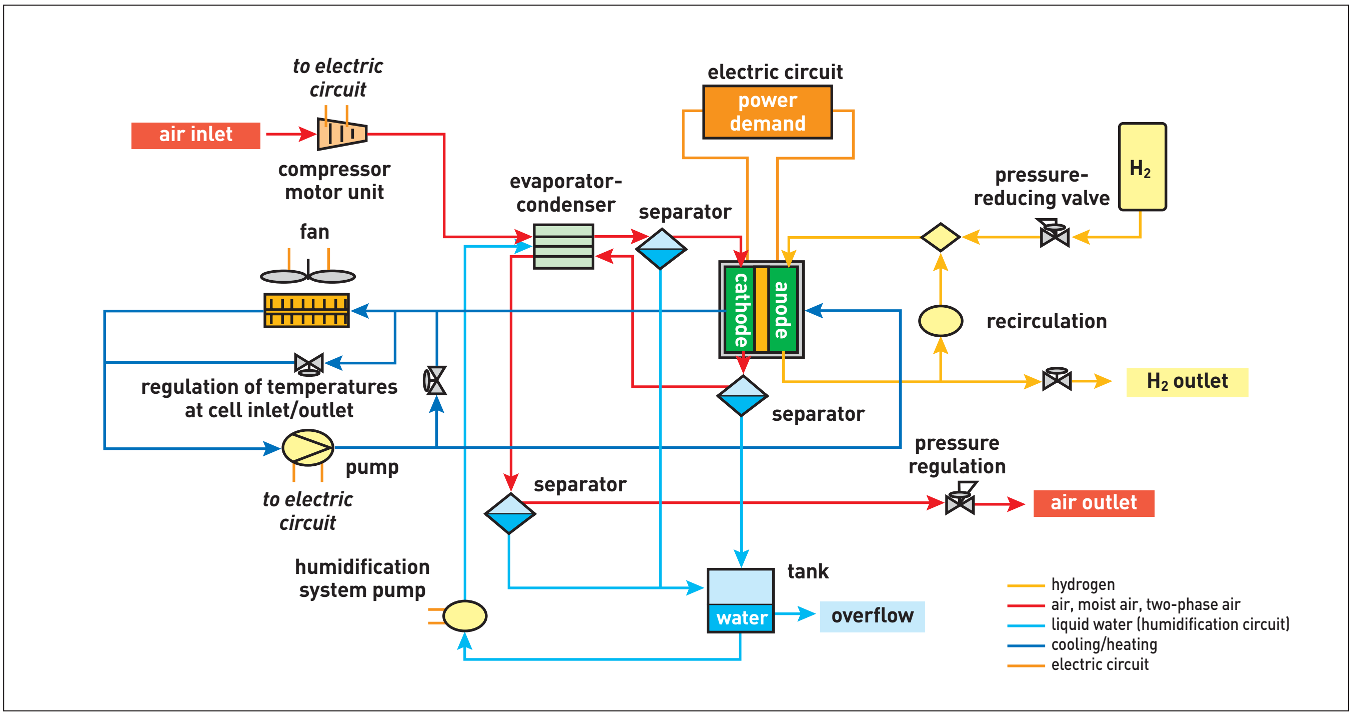

Figure 1.
Simplified schematic of the electric generator unit in the Hydro-Gen vehicle.

the other hand, the constraints specific to each application increasingly result in distinctive system designs, and even development of specific basic components: MEA (membrane-electrode assembly), bipolar plate, but equally compressors and gas-feed humidification system. The nature of the primary fuel used, and the system thermal rejection or electric signal conditioning requirements all have a decisive impact on the system.

This paper aims to present an outline of the approach used for the definition of a PEM fuel-cell system, as illustrated by the automotive application.

Targets, basis, and initial givens

The automotive application is the most competitive, and one where constraints are most stringent. In the absence of **hybridization**, target efficiencies for the system are 50% at nominal power rating (50-80 **kW**), and 60% at a quarter of that power rating. Target longevity is 5,000 hours, and 10 years. Requirements more specifically related to the application essentially concern system compactness (650 W/L, 650 W/kg), cost (€45/kW), and ability to withstand extreme temperatures, ranging from - 20 °C to + 45 °C, for all operating phases, including start-up.

Manufacturers are all agreed, nowadays, that the most promising onboard fuel is pure **hydrogen**. Storage of hydrogen in compressed form does indeed make for simpler design. Figure 1 shows the simplified schematic of a typical electric generator unit from the period 1995-2000, the system for the Hydro-Gen vehicle. (1)

This may be broken down into five sub-systems: hydrogen supply for the fuel cell, fuel cell air supply, the fuel cell stack itself, electric power management, and humidification and thermal management. Sizing of such a system is subject to a tradeoff between application-dictated requirements (performance and functionalities) and the functional relationships between the various parts of the system.

Knowledge of a number of initial givens is indispensable for the sizing of the fuel cell stack, the central component in the system. The basic givens are system nominal power and target energy efficiency. A **stoichiometric** ratio must also be set for hydrogen (ratio between the amounts of hydrogen going into the system, and hydrogen consumed); this is commonly close to unity. Finally, estimation of parasitic power, i.e. power consumed, in particular, by the compressor, is required.

Fuel cell stack sizing

From these data, fuel cell nominal power and energy efficiency may be calculated. The latter is directly proportional to **basic cell** voltage, this now forming the datum point from which sizing may proceed. The "electric power management" part specifies the voltage range the fuel cell stack must keep to, which dictates the number of basic cells and the current to be supplied. From the values set for the current, basic cell surface area may be determined, depending on MEA performance. For this purpose, the value must be known of the **current density** delivered by the MEA, for the target efficiency. This value depends on operating conditions: pressure, humidity, input stoichiometric ratio at the **anode** and **cathode**, temperature (see Figure 2). Basic cell surface area and number of basic cells form the basis for fuel cell sizing, in terms of mass, volume, and cost. The choice of technology for the fabrication of components, such as the bipolar plate, derives from this, to comply with mass and volume constraints. Achieving performance levels such as 1 kW/kg and 1 kW/L is not compatible with certain technologies. For instance, at current MEA performance, use of **graphite** or filled **polymer** allows performance levels to

(1) Funded by the European Union, the Hydro-Gen Program (1996-2001) brought together PSA Peugeot Citroën, Air Liquide, Renault, Nuvera, Solvay and CEA for the purposes of constructing a fuel-cell vehicle.

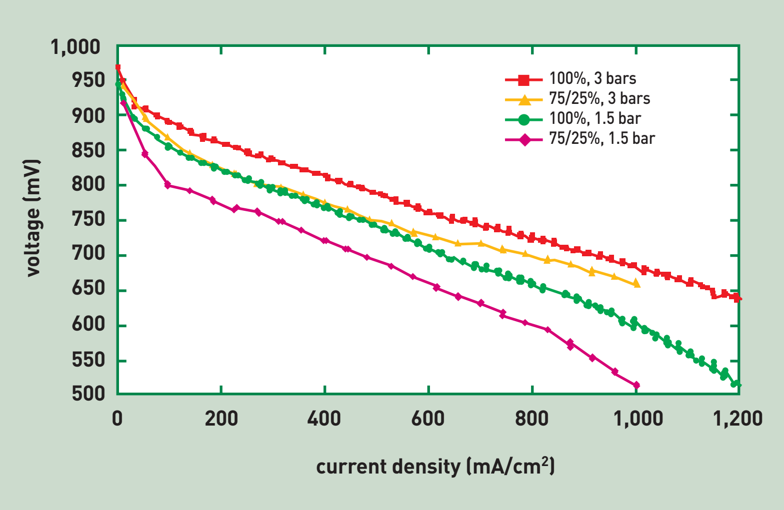

Figure 2.
Current cell-core performance, for various values of pressure (in bars) and gas-feed moisture content at the anode and cathode (%), shown in terms of output voltage as a function of current density.

be expected close to 0.6-0.8 kW/kg and 1 kW/L, while metal technologies (thin stamped sheet, or foam) offer the capability of yielding fuel cell stacks attaining over 1 kW/kg, and 1.5-2 kW/L. Each of these technologies presents further benefits and disadvantages, as well as being at varying stages of development (see *Innovative concepts for bipolar plates*).

It should be noted that this sizing is dependent on chosen operating conditions, namely pressure, temperature, gas flow rates, and input gas moisture content. These choices determine the makeup of the various constituent parts of the system, which in turn are subjected to system constraints. Thus, for the automotive application, choice of basic cell temperature is dependent on the vehicle's radiator heat rejection capacity.

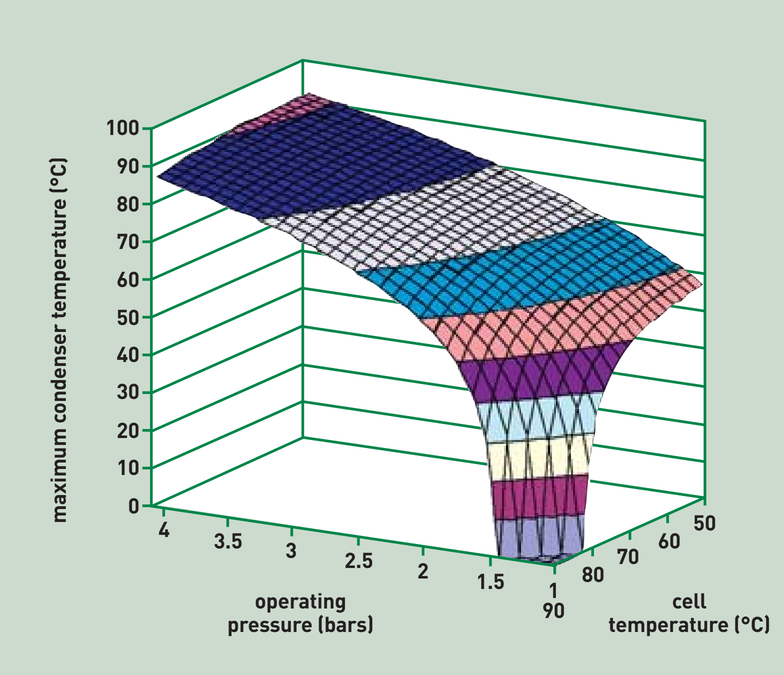

Figure 3.
Condenser temperature, expressed as a function of cell temperature and the operating pressure beyond which water balance shows a deficit.

Sizing of the cooling system

Compared to a heat-engine vehicle, a fuel-cell vehicle must reject a far greater amount of heat via its radiator. Indeed, heat rejection via exhaust gases, and by radiation accounts for over half of the heat generated, in the heat engine case, whereas this is negligible in the fuel cell case. Further, the coolant is at a higher temperature for a heat engine (120 °C), enhancing radiator efficiency. Sizing of the cooling system is thus a key point in system design. Parameters impinging on this are the temperature differential between the coolant and the atmosphere, radiator section, fan power, coolant fluid flow rate, and fuel cell efficiency.

The choice of fuel cell temperature, directly linked to the coolant temperature, largely determines sizing. In the automotive application case, outside atmospheric temperature may be high - up to 40 °C, or even 50 °C - entailing high temperatures for the coolant, and hence for the fuel cell. On the other hand, the value for cell temperature is restricted to 80-90 °C, for a number of reasons, including long-term MEA behavior, or the difficulty of ensuring gas-feed humidification at such temperatures. There is a further requirement to increase radiator size (by a factor 1.3) and fan power (by a factor 4).

In the context of applications other than the personal vehicle case, choice of a lower cell temperature becomes a possibility (typically, 60 °C), offset by oversized cooling components. This type of functioning is considered for stationary systems, for which volume and mass are less of an issue. The benefit then consists in a simplified humidification system.

For all cases, however, it is feasible to reduce the amount of rejected heat, by raising the fuel cell efficiency, through cell oversizing, a process involving a down side in terms of system mass, volume, and cost.

Water management sizing

To ensure a level of water content in the membranes compatible with good **ion conductivity**, it is essential to effect humidification of the gases at the fuel cell inlet. As a default option, it is possible to choose to saturate the gases with water, however, the amount of heat required for evaporation can become considerable, at high temperature and low pressure, even exceeding the amount of heat generated by the fuel cell. Added to this is the issue of water management, this consisting in recovering at the cell outlet at least as much water as is required to humidify the gases going in. The amount of water recovered is a function of the amount of water generated, and condenser temperature. Such a functioning has its limitations at high temperature and low pressure (see Figure 3). The condenser temperature required to sustain water self-sufficiency for the system thus becomes unattainable in some conditions.

However, current MEA allow getting away from this constraint, through structural optimization, allowing operation with partially humidified gases. Nevertheless, the foregoing argument still stands.

Sizing of air supply

Water management and the quest for high cell efficiency provide incitements to operate at high pressure.

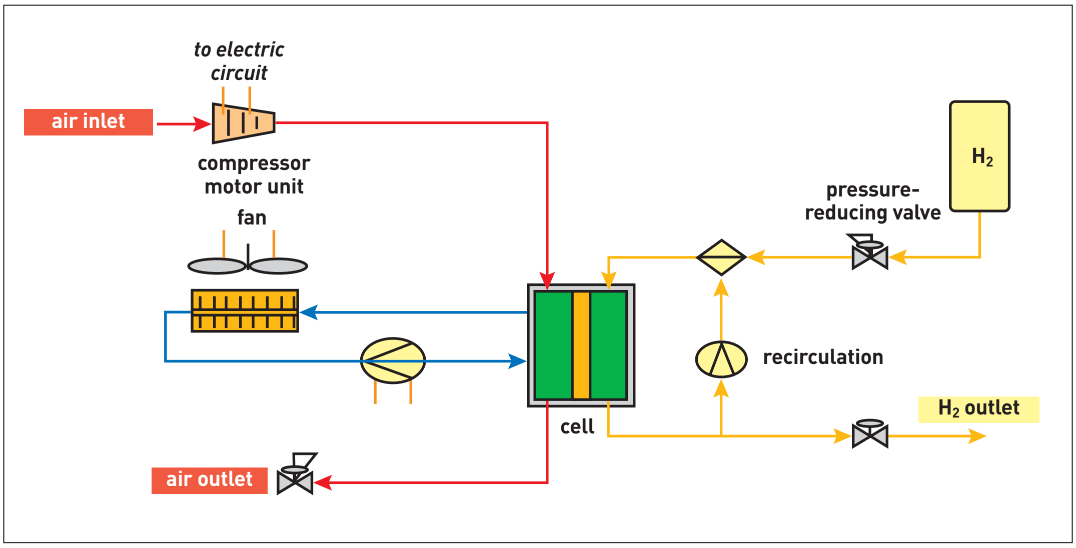

Figure 4.
Schematic of a simplified system, with no expander.

The counterpart for such a choice is increased energy loss from air compression, this being a function of cell working pressure and stoichiometric ratio for air. As an example, this accounts for more than 10% of cell gross power at 2 **bars**, reducing, or even nullifying, the gain achieved in terms of cell voltage. One way of lowering this parasitic energy expenditure is through recovery of part of the mechanical energy contained in the outgoing airflow at the cell outlet, by means of an expander. This option increases system complexity, and complicates system management.

Towards a degree of simplification

Current trends for the various system components are pointing to greatly simplified systems. Improved MEA performance, particularly at low humidification rates, fuel cell stack design allowing easier high-temperature, low-pressure operation, and advances in air compression are the paths making for such simplification. The ideal system could that shown in Figure 4.

Systems for stationary applications are subjected to less stringent constraints, in particular as regard compactness and heat rejection. Low-temperature operation, made possible owing to the absence of compactness requirements, makes water management issues, and hence choice of operating pressure, less fraught with difficulty. The fuel cell stack, and equally the other components in the system may then be different from those for the vehicle application, with the ability, for instance, to use air cooling, rather than liquid cooling. The overall definition procedure, however, remains unchanged.

For all cases, the issue not taken into account in this sizing is that of durability, in particular for MEA. It is to be expected that this constraint will lead to tradeoff modifications.

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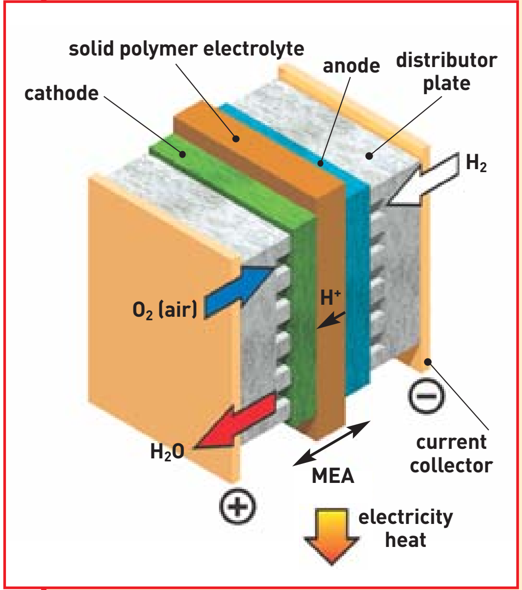
PSA Peugeot Citroën - Communications Directorate

The new Quark demonstrator, designed by Peugeot, a two-seater vehicle in which a fuel cell complements the electric power supplied by a nickel-metal hydride battery. Integration of the fuel cell required much investigation to achieve system simplification and "compacting."

C

How does a fuel cell work?



Operating principle of the fuel cell: the example of the proton-exchange membrane fuel cell. MEA stands for membrane-electrode assembly.

The fuel cell is based on a principle discovered quite some time ago, since it was in 1839 that Sir William Grove constructed the first electrochemical cell working with hydrogen as its fuel, thus demonstrating the ability to generate electric current through direct conversion of the fuel's chemical energy. Since the fuel cell has the special characteristic of using two gases - hydrogen H_2 and oxygen O_2 - as its electrochemical couple, the oxidation-reduction reactions occurring inside the fuel cell are particularly simple. The reaction takes place inside a structure (the basic electrochemical cell), consisting essentially in two electrodes (the anode and cathode), separated by an electrolyte, i.e. a material that lets ions through. The electrodes employ catalysts, to activate, on the one side, the hydrogen oxidation reaction, and, on the other, the oxygen reduction reaction.

In the case of an acid-electrolyte cell (or proton exchange membrane fuel cell), the hydrogen at the anode is dissociated into protons (or hydrogen ions H^+) and electrons, in accordance with the oxidation reaction: $H_2 \rightarrow 2\ H^+ + 2\ e^-$. At the cathode, the oxygen, the electrons and the protons recombine to yield water: $2\ H^+ + 1/2\ O_2 + 2\ e^- \rightarrow H_2O$. The principle of the fuel cell is thus the converse of that of water electrolysis. The thermodynamic potential for such an electrochemical cell, consequently, stands at around 1.23 volt (V). However, in practice, the cell exhibits a voltage of about 0.6 V for current densities of 0.6-0.8 A/cm^2. The efficiency of such a fuel cell is thus equal to about 50%, the energy dissipated naturally being so dissipated in the form of heat.