



Hybridization, Diagnostic and Prognostic of Proton Exchange Membrane Fuel Cells

Durability and Reliability

Samir Jemeï



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Alain Dollet

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Foreword

When reflecting on what our energy future should be, specialists in the field use various terms: abundance, sustainability, renewable within human life span, environmental friendliness, accessibility to all in every corner of the planet and multiplicity of application contexts. Hydrogen meets all these requirements. It is indeed the most abundant element in the universe (75% in mass and 92% in number of atoms). It would then seem logical and even obvious to consider it an interesting component in our future energy mix. Unfortunately, since it is rarely found on Earth in its dihydrogen form, it needs to be produced. The hydrogen thus obtained can be characterized as a “hydrogen energy vector”, which forms a natural duality with the electricity vector (passages from one vector to another are obtained either by water electrolysis or by using a hydrogen fuel cell). Because of this duality, its use is rendered particularly easy, while introducing an intrinsic capacity to limit the environmental impact of the full energy chain (indeed, hydrogen can and should be produced without fossil fuels). The commonly used term is hydrogen-energy, in which the suffix clearly highlights the goal to be obtained.

In this rapidly evolving energy context, disruptive technologies (among which is hydrogen-energy) are worth being studied in depth. In this book, we rightly propose a systemic view of technological, economic and social stakes related to this energy vector. Owing to his unique experience of approximately 20 years in this field, Dr Samir Jemei goes beyond this scope. He draws on this experience to identify the major scientific challenges to be met in order to bring hydrogen fuel cells to the level of technological maturity, which would enable them to become fully competitive commercial products.

Therefore, this work offers interested readers an in-depth view on these subjects, focusing on two major issues: hybridization of hydrogen power generators and identification of their (current and future) state of health. The results of numerous scientific studies conducted in recent years are thus summarized here. The advantages presented by using advanced signal processing methods for real-time control of energy flows in hydrogen fuel cells is particularly worth noting, together with artificial intelligence-based approaches for the state of health diagnosis and remaining lifetime prognosis for these electrochemical generators.

Despite the complexity inherent in the studied systems, this book provides an astonishingly didactic value for the layperson. The reader can thus become progressively acquainted with the stakes, methods and adopted strategies (What are the study requirements? What are the specific constraints associated with the hydrogen fuel cell? What are the various possible approaches? What are the principles? What measurements enable a choice? What are the expected results?). The very rich bibliography provides readers with the opportunity to expand their exploration of a specific field of study. A further teaching benefit stems from the approaches used, which are systematically evaluated, thanks to experimental data (generated by experiments conducted on hydrogen fuel cell testing bench or on full experimental systems (particularly hydrogen-powered electric vehicles)).

With a skillful combination of scientific theory and industrial practice, this book is an essential reference for all the engineers and researchers working in the field of integration of hydrogen fuel cell systems, either in environmentally friendly stationary applications or in clean transport.

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May 2018

Introduction

I.1. Subject matter

Global emissions of greenhouse gases covered by the Kyoto protocol reached approximately 49 billion tonnes of CO₂ equivalent in 2010. They increased by 80% between 1970 and 2010 and by 30% since 1990 [FRA 15]. The large majority of these emissions are obviously related to the combustion of fossil energies. The resulting pollution and the irreversible depletion of fossil resources should stimulate a reflection on our global energy system, which seems on its last legs. Many measures that have been taken through various programs, such as sustainable development, Horizon 2020 and even COP21, aim to decrease greenhouse gas emissions, reduce energy consumption, stabilize global warming, diversify primary energy sources or even develop renewable energies. Taking into account these elements forces the system into a new energy transition and tends to increase the share of renewable energies in the energy mix.

Power generation using wind and solar energies is reaching maturity, with ever-increasing production. Nevertheless, irregularity in the availability of these renewable energies gives rise to many problems in terms of power grid management. Managing a grid that day by day involves slightly more intermittent energies requires availability of reserve storing and release in order to cope with the differences between supply and demand in a given territory. It is indeed important to be able to deal with various time scales: daily (day/night), weekly (weekend/weekdays) and seasonal (summer/winter). Yet, it proves that hydrogen is ideal for storing this renewable energy. It can be produced using water electrolysis, which is itself

fed by renewable energies. The resulting gas is then stored under various forms in order to be used at the appropriate time. Hydrogen can be combined with natural gas in existing networks, can be directly used in the industry or can even be used to again generate electric power by means of a fuel cell.

Since the fuel cell (FC) is at the center of the research activities presented in this book, it is worth our full attention. Indeed, this electrochemical generator converts the chemical energy of a reaction between a fuel (hydrogen) and an oxidizer (oxygen in the air) to generate electricity, heat and water. It can be used for stationary or nomadic applications or in transport. Moreover, in order to decarbonize the transport, the use of the hydrogen energy vector in combination with a fuel cell becomes an increasingly mature solution. As a proof, many global car manufacturers are manufacturing and marketing their own fuel cell vehicles. The most recent example is that of the Japanese manufacturer Toyota, which sold approximately 1,000 units in 6 months and is expecting to reach 30,000 fuel cell vehicles per year from 2020.

In Japan, except for stationary applications, more than 200,000 fuel cell systems have so far been installed in private homes to generate electricity and heat.

Hydrogen can be used in all sectors to store intermittent energy, supply mobile applications, meet the needs of an isolated site, which is disconnected from the power grid, propel a car, railway, naval or space flight transportation means and so on. In 1874, in his book *The Mysterious Island*, Jules Verne had predicted without an artificial intelligence algorithm that: “*water will one day be employed as fuel, that hydrogen and oxygen which constitute it, used singly or together, will furnish an inexhaustible source of heat and light, of an intensity of which coal is not capable*” [VER 74]. Half a century later, the rise of a hydrogen society is conceivable.

This book essentially deals with the problems related to fuel cell technology for energy generation. Even though the FC has experienced much progress in the last 20 years, the technical challenges have not yet been fully solved. The research work described in this book falls within this context, drawing on the works that relate to electrical engineering and, to a lesser extent, to the automation field.

1.2. Chapter breakdown

The works presented are centered on three axes organized in four chapters.

The first chapter starts with a brief overview of the current energy model. A review of the solutions that could enable the decarbonization of the energy mix is then provided. Among these solutions, a proposed focus is on the hydrogen vector, which is closely linked with the fuel cell generator. This chapter ends with the presentation of the fuel cell and its applications. Potential applications for transport and for the stationary field are highlighted. Finally, advantages and drawbacks of this technology are described. The FC presents problems related to cost, efficiency, lifetime, integration and hydrogen storage, but it has the advantage of releasing few or no pollutants, being silent, having high energy efficiency and offering substantial lifetime and reduced cost. Nevertheless, the FC is a multi-physical generator that becomes difficult to understand when considered with its auxiliaries.

The second chapter presents various existing types of FCs and identifies two technologies, namely the PEMFC (proton exchange membrane fuel cell) and the SOFC (solid oxide fuel cell), which are considered to be the most promising solutions for transportation and stationary applications. Regardless of the technology used, many auxiliaries are needed to operate the FC and it is important to control the whole system. Hence, all the auxiliaries are described and particular attention is given to the compressor unit that allows the air supply of the PEMFC, which is one of the largest energy consumers of the system (about 10–15%). With regard to the SOFC, which mainly differs from the PEMFC by its operating temperature (above 800°C), an integrated power generator is presented. For both technologies, experimental implementation is a key point, as it facilitates the understanding of how the FC and its system operate. These works can be considered the first axis, on which the research activities needed to obtain an augmented durability FC system rely. The second axis deals with the energy optimization of various hybrid energy sources, which enables an increase in FC lifetime. Finally, the third axis is dedicated to FC diagnosis and prognosis, which moreover enable the expansion of lifetime and an increase in generator reliability.

Thus, the third chapter deals with the hybridization of energy sources. If a transport application is considered, it is important to hybridize the FC with other sources, as the overall dynamics of the FC system is rather slow and causes difficulties in the case of rapid transients within the mission. Furthermore, in order to reduce the overall energy consumption of a vehicle, braking energy should be recovered, which is impossible with a single FC, as it is a non-reversible source. Finally, hybridization of this FC with energy-storing devices should be considered, in order to obtain high overall efficiencies. In such hybrid systems, an energy management strategy must be developed so that power demands are allocated to the right sources, at the right times, depending on the mission profile to be followed. Two approaches are proposed in this chapter. The first approach, which relies essentially on the wavelet transform combined with neural networks, enables the development of online energy management applied to a heavy hybrid vehicle composed essentially of a FC system, accumulators and ultracapacitors. It is worth noting that this energy distribution takes into account the frequency ranges of various sources using only the current and previous data of a univariate signal of the vehicle power demand. This will eventually allow real-time implementation as well as an increase in FC lifetime. The second approach relies on type-2 fuzzy logic and genetic algorithms. This solution is applied to a hybrid locomotive, composed of accumulators, ultracapacitors and a diesel engine, in order to minimize fuel consumption. The results show that the characteristics and dynamics of various sources have been taken into account thanks to artificial intelligence tools. High-performance energy management tools are proposed here, allowing optimal use of sources. Nevertheless, our efforts should focus on FC lifetime and reliability, in order to have a hybrid system with even higher performances.

For this purpose, FC diagnosis and prognosis have been developed in the fourth chapter. These two disciplines should enable us to reach our objective of increasing the lifetime and reliability of the FC. Diagnosis methods are used to identify the origin of failure and to determine the FC state of health, thus allowing for decision-making that ensures proper operation of the generator. PHM (Prognostics & Health Management) helps to predict the evolution of FC behavior in order to estimate a future failure. There are many approaches to the development of diagnosis and prognostic tools. Nevertheless, in order to find effective solutions, first it is important to have in-depth knowledge of the degradation mechanisms of the FC and its

system. Thus, in the first part of Chapter 4, degradation mechanisms and the failures that the FC and its system may present are reviewed. In the second part, two diagnosis methods are presented. The first one (data-based) is a supervised classification method called “k-nearest neighbors”. The second (signal-based) relies on a wavelet transform approach. These approaches have in particular enabled the diagnosis of various FC system failures that may occur with good classification rates reaching 90%. As will be shown, it is also possible to estimate the FC state of health using the wavelet transform coupled with energy indicators. Finally, in the last part of this chapter, methodologies related to the data-based prognostic are developed. The tools used to monitor the state of health and estimate the future behavior of the FC are based on neural networks. The first works are carried out using the ANFIS (Adaptive Neuro-Fuzzy Inference System). This model enables us to predict the behavior of voltage evolution. Nevertheless, the learning bases prove to be substantial. To reduce the required data and achieve better prediction performances, we have subsequently used the ESNs (echo state networks), which are new systems of neural networks. The conventional neural networks are often costly in terms of computation time due to algorithmic complexity. For the ESNs, algorithmic complexity is replaced by structural complexity, so that the learning phase is faster than that of conventional neural networks. Thanks to the use of ESNs of genetic algorithms and the wavelet transform, the results obtained are very encouraging. Indeed, they allow us to predict cell voltages until the end of life of the FC with an error below 10%.

Fuel Cells: the Path Towards Hydrogen Revolution

1.1. Introduction

In the 21st Century, it has become nearly impossible to do anything without means of transportation, communication or energy generation, which are expected to be increasingly efficient. These three concepts are present in many fields of activity of everyday life, such as telecommunications, information technology (thermal and/or electric) and energy supply for domestic or industrial use or for transportation (private, railway, maritime or air). These various technological means must have very high efficiency and release a minimum amount of pollutants, in order to increase the availability of our fossil energy resources and preserve our planet. To date, more than 80% of the primary energy supply relies on fossil fuels [BAD 13], which leads us, on the one hand, to a supply breakdown in the very near future and, on the other hand, to an environmental catastrophe. Indeed, the most significant and immediate risk relates to the climate change caused by the emissions of greenhouse gases due to human activities. The use of fossil fuels generates CO₂, which is the main man-made greenhouse gas contributing to global warming. In order to avoid an increase in Earth's average surface temperature by more than 2°C, human CO₂ emissions should be halved by 2050 globally [INT 15]. For this purpose, the European Union has set various objectives in terms of climate change and energy until 2020. It is the “three times twenty” rule that should enable the start of genuine energy transition:

– decrease in greenhouse gas emissions by 20% (even by 30%, if possible) compared to 1990;

- increase in energy efficiency by 20%;
- up to 20% use of energy from renewable sources for primary energy supply.

Firstly, this chapter will provide a brief overview of global energy, going through various possibilities for the future.

Secondly, the focus will be on the hydrogen vector and on the need to store it.

Finally, the fuel cell generator and its applications will be presented.

1.2. Energy: a global view

Global energy consumption has rapidly increased in the recent past, relying essentially on fossil energies. This explains why significant progress has been made over the past few years by developing cleaner and more efficient production technologies. Nevertheless, agreed efforts are not sufficient to avoid a 2°C increase in the Earth’s average surface temperature. This requires an “energy mix” that will enable the preservation of resources and the environment, while at the same time meeting the demand.

1.2.1. *An obsolete energy model*

Energy demand is steadily increasing due to the requirement to meet the energy needs of increasing population. As previously mentioned, fossil energy reserves will be depleted and alternative energy sources need to be found. It should be recalled that in 2010, 80% of the global primary energy generation relied on fossil energies, 10% on hydroelectric energy, 6% on nuclear energy, 2% on biomass energy and only 1% on other renewable energies [INT 12]. Unless measures are taken, the carbon dioxide content in the atmosphere could increase by 30% in 20 years, while focusing on the development of other energy resources that generate less carbon dioxide waste, which could help us reverse the trend and decrease the emissions by 30% by 2035 (Figure 1.1). Examples of resources or technologies that release the lowest rates of CO₂ include solar, wind, hydraulic and nuclear energy (Figure 1.2). It is thus essential to propose a combination of renewable energies that can satisfy our various preoccupations and, in particular, to further develop decarbonized energies.

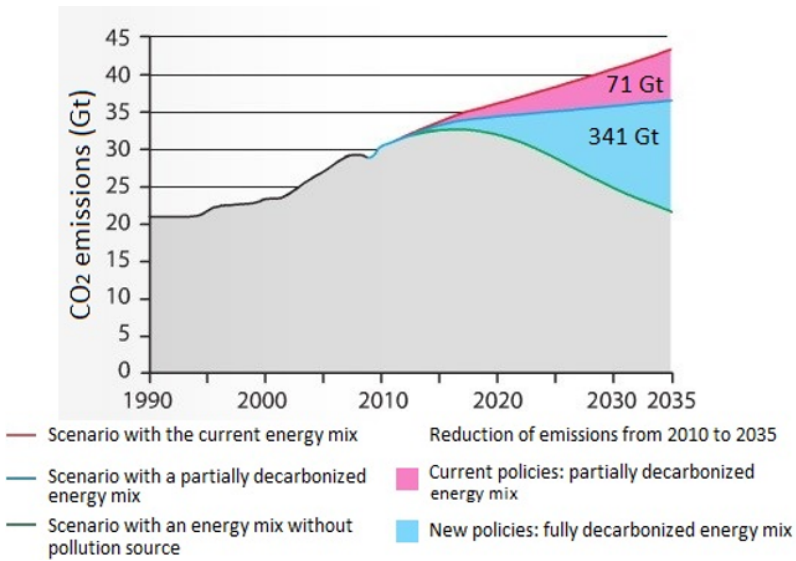


Figure 1.1. Scenarios of the evolution of CO₂ emissions [INT 11].
For a color version of this figure, see www.iste.co.uk/jemei/cells.zip

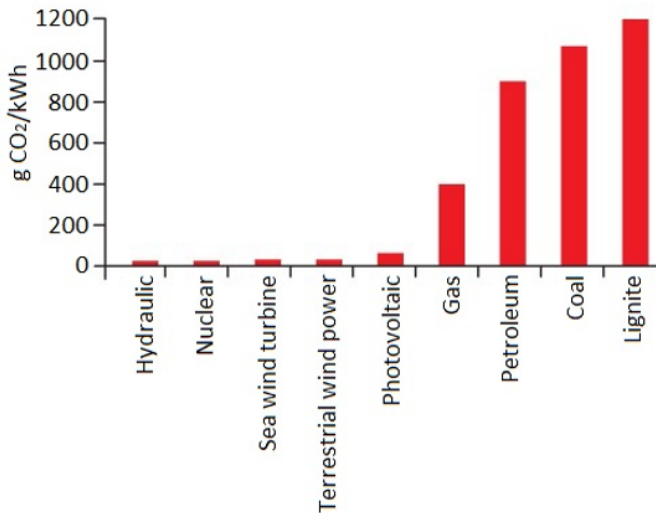


Figure 1.2. Direct emissions + analysis of CO₂ life cycle by energy source [BER 14]

1.2.2. Solutions for the energy mix decarbonization

Nowadays, there are many possibilities (nuclear, renewable, etc.) that enable the replacement of fossil energies by renewable energies. Nevertheless, various possible replacement solutions should be considered in terms of competitiveness, economic viability, technical feasibility and social acceptability. In this book, we focus exclusively on solutions relying on renewable energies.

“Renewable energies are inexhaustible and they rely mainly on solar energy.”

In 2012, global renewable production of electricity (including pump and turbine power generation) reached 4,699.2 TWh, clearly crossing the threshold of 20% of global electricity production (20.8%). Renewable electricity can be generated from six distinct sources. Hydroelectricity (including pump and turbine power plants) is the first among them, with a 78% contribution in 2012. Wind energy, which from 2009 has become the second common source of renewable energy, and now represents 11.4% of all renewable energy. It outperforms biomass, which covers solid biomass, liquid biomass, biogas and renewable domestic waste (6.9%). It is followed by solar energy (2.2%), which includes photovoltaic power plants and solar steam plants, geothermal energy (1.5%) and ocean energies (0.01%), which are still in the demonstration phase [OBS 13]. A basic review of these various sources is provided below [FRE 13].

Hydroelectric energy: hydroelectricity recovers the motive power of streams, waterfalls and tides and converts it into electricity. There are two types of installations: “run of the river” hydroelectric installations, which run all or part of a stream flow through a turbine continuously, and those that require water reserves (“feed of water” or “lake”). These two types of installations require dams, which are much more important for the second category (“large dams”).

Wind energy: wind turbines enable the conversion of wind energy into electric energy. Thanks to the technological progress, mass production and accumulated experience, wind energy should become the largest contributor to electricity generation in the decades to come.

Biomass: there is a considerable difference between biomass and other renewable energy sources, to the extent that it takes the form of a fuel that can be stored and used to generate electricity as needed, similarly to fossil fuels. Nevertheless, biomass is often limited by the energy density of the stored fuel. The following are the three basic technologies for converting biomass energy into electricity, the primary fuel being solid-state biomass:

- direct combustion;
- gasification;
- pyrolysis.

Solar energy: solar power plants convert solar radiation into electricity or heat, depending on the technology employed.

Photovoltaic solar energy uses photovoltaic modules to generate electricity, which is then fed into power grids.

Solar thermal energy generates heat, which can be used for domestic heating or hot water generation.

Finally, thermodynamic solar energy generates electricity by heat production.

Geothermal energy: geothermal energy or “heat of the Earth” covers all the applications, enabling the recovery of the heat contained in the subsoil or in the water table (the higher the temperature of the ground and of the underground water, the closer one gets to the center of the Earth). Depending on the application, the calories thus recovered serve to generate heat and/or cold or electricity [NOT 17].

Ocean energies: ocean energy conversion includes all the technologies that enable energy generation, particularly electricity, based on the various resources of the ocean environment, such as swell, currents, tides, temperature gradient and wind (sea wind turbines) [NOT 17].

The European objective, which sets the share of renewable energies at 20% (23% for France) in the final gross consumption of energy until 2020, involves a strong development of intermittent sources such as wind or solar energies, whose generation profiles are sometimes out of phase with the profiles of electric energy consumption [GRH 16].

It is therefore essential to develop flexibility vectors that optimize the connection between energy generation and consumption. The hydrogen vector is one of the relevant candidates to be used as a flexibility solution.

1.3. Hydrogen vector

Hydrogen does not exist on Earth in its natural state. This element is the lightest and one of the most abundant in nature. It is necessarily linked to other elements such as dioxygen or carbon. Hydrogen can be generated by extracting and isolating the molecule. Water electrolysis is another solution. The dissociation of water molecule requires the same amount of energy that is recovered when hydrogen and oxygen are recombined. Hence, hydrogen is not a resource, but an energy vector.

1.3.1. Hydrogen production

Hydrogen can essentially be produced from three large sources, namely fossil, nuclear and renewable energies. Nowadays, half of the global hydrogen production results from methane reforming, which is the most economic source, and the other half is obtained from coal or petroleum. Electrolysis, which is an expensive technique, accounts for only 4% of the global hydrogen production, particularly when very pure hydrogen is needed. The supply of refineries and the fabrication of ammonia constitute 85% of the outputs [BAD 13].

1.3.1.1. From fossil energies

Hydrogen production in industrial applications relies on methane (CH₄), which is the main component of natural gas. The method used for this purpose is reforming. It is a chemical reaction that breaks the hydrocarbon molecules under the action of heat and releases hydrogen (dihydrogen, more precisely). There are two types of reforming:

– *Steam reforming* of natural gas is the most common process. Mixing methane and water vapor at high temperature (about 850°C) triggers the endothermic reaction (equation [1.1]):



with $\Delta H^0 = 252.3 \text{ kJ.mol}^{-1}$