Hybrid fuel cell system degradation modeling methods: A comprehensive review

Loïc Vichard, Nadia Yousfi Steiner, Noureddine Zerhouni, Daniel Hissel

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L. Vichard[a], N. Yousfi Steiner[a], N. Zerhouni[b], D. Hissel[a]

[a]FEMTO-ST Institute, FCLAB, Univ. Bourgogne Franche-Comté, CNRS, Belfort, France

[b]FEMTO-ST Institute, FCLAB, Univ. Bourgogne Franche-Comté, ENSMM, CNRS, Belfort, France

Abstract:

Last years, the fuel cell has become well-known as an efficient and clean energy converter being a potential alternative to internal combustion engines. However, despite being very promising, the durability of those systems is still a bottleneck. Most of the time, a fuel cell is integrated in a hybrid system which considers the fuel cell stack, the battery, and the balance of plant. To keep improving the durability of such a system, diagnostic and prognostic tools are of particular importance and to implement such tools, modelling the system is a mandatory step. The purpose of this paper is to propose a critical review of the existing methods to model all elements of a hybrid fuel cell system according to operating conditions and degradation. In this review, interactions and major degradation mechanisms occurring at all components will be presented and the physics-based models, data-driven and hybrid models of these components reviewed. Finally, methods will be discussed, and advantages and drawbacks will be summarized.

1 Introduction

1.1 Generalities

In the last years, the popularity of hydrogen has been highly growing up as it plays an increasing role in a necessary energy transition. Proton Exchange Membrane Fuel Cell (PEMFC) is regarded as a technology of great potential because of the high energy density, high conversion efficiency, low operating temperature and low greenhouse gases emission [1]. Nevertheless, the durability remains a limitation to compete with traditional combustion engine and achieve a worldwide commercialization [2,3]. Most of the time, a fuel cell is integrated in a hybrid system which considers the fuel cell stack, the battery, and the balance of plant.

Integrated into a vehicle, fuel cell systems are subject to rough operating conditions [4,5] such as dynamic load variations [6][7], air impurities, temperature and humidity variations, cold ambient temperature [8], vibrations and shocks [9] which can accelerate the degradation of the system components.

For these reasons, elaborate diagnostic and prognostic tools based on system model according to different configurations and energy management strategies is of particular importance in order to estimate and improve performance with regards to reliability and durability. However, setting up reliable diagnostic and prognostic tools relies on a good understanding and mastering of faults operating conditions and degradation mechanisms at system level.

Based on historical data describing the performance evolution, a degradation model allows to reproduce, analyze, and forecast the dynamic behavior, as well as the state of health (SOH) of the considered system. Different approaches have been proposed in the literature, which can be classified according to three major categories: physics-based models, data-driven models, and hybrid models.

The purpose of this paper is to provide a comprehensive and critical review of the existing methods to model all elements of a hybrid fuel cell system considering performance degradation due to operating conditions and aging of components. The modeling approaches will be identified and selected according to the available knowledge about the system, and ability to be implemented real-time, and the objective of the modeling, namely design, optimization, durability, and reliability.

1.2 A hybrid fuel cell system

As illustrated in Figure 1, a hybrid fuel cell system is composed of the stack which is the heart of the complete system and the associated balance of plant (BoP) that includes a heat exchanger, air compressor, H_2 tank, cooling system, humidifier, power converters, etc. As a hybrid fuel cell system, it also integrates other power sources such as batteries and/or supercapacitors. In this study supercapacitors will not be considered. The illustrated architecture is one of the most common existing in actual applications, but it can vary. Indeed, there are architectures which can integrate a DC converter as in output of the battery. Also, Battery Management System (BMS) and Fuel Cell Control Unit (FCCU) can be separated into two different controllers. Hybrid systems can also integrate multi-stack connected in parallel.

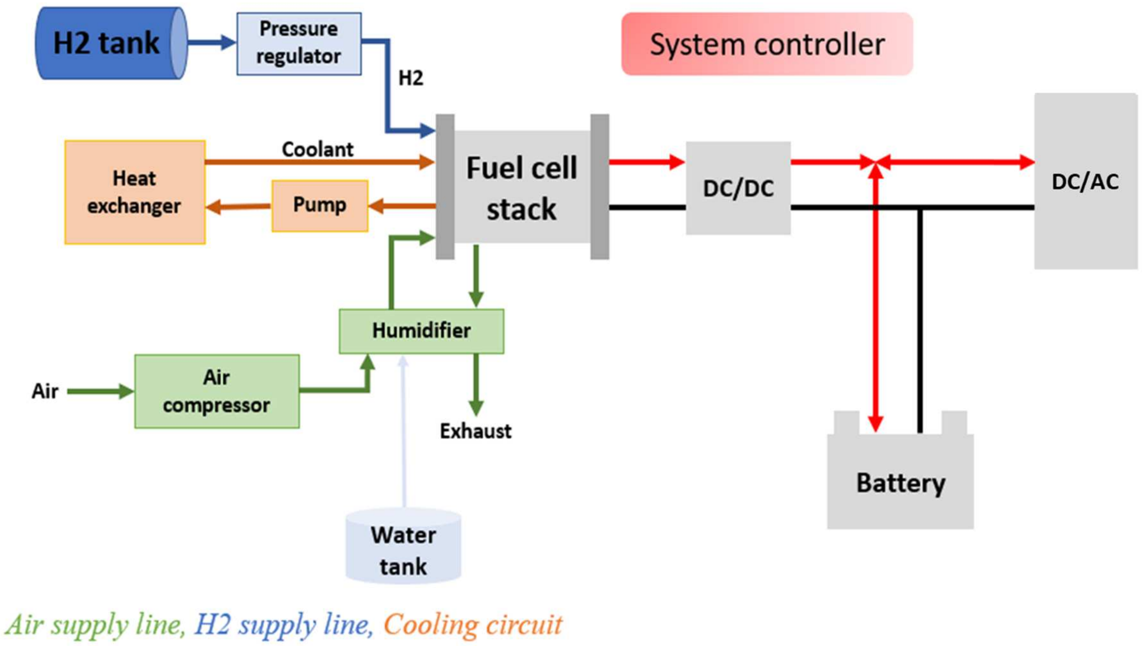

Figure 1: schematic of a typical hybrid fuel cell system and balance of plants

It is worth noting that this Figure highlights the architecture of a hybrid system in the case of a closed cathode water cooled fuel cell stack. Indeed, in the case of an open-cathode air cooled fuel cell stack, only the hydrogen line remains similar as the stack cooling and cathode air supply are provided by fans [10].

Considering the architecture of a hybrid fuel cell system highlighted in Figure 1, there are many interactions between the components of the system. The hydrogen line provides the fuel (hydrogen) to the fuel cell stack anode required to start the chemical reactions. The input hydrogen pressure is set by a pressure regulator. The air line provides the oxidant (air) to the fuel cell stack cathode required to start the chemical reactions. A compressor is used to control the air mass flow at the input of the cathode side and the air pressure at the output. A passive humidifier is integrated between the air compressor and the fuel cell stack in order to add water content to the oxidant. A humidified air (oxidant) is of particular importance for operation of the fuel cell stack in the best conditions without what the membranes could tend to dry out and generate cracks and pinholes within the electrolyte.

The cooling system allows to control the temperature inside the fuel cell. Chemical reactions occurring within a fuel cell stack are exothermic. Over-temperature will lead to a destruction of the components. Therefore, fans coupled with a heat exchanger and a water circuit are controlled in order to regulate this temperature. The power converter ensures the electric power conversion from the output of the fuel cell stack to the DC bus.

The role of the battery is to ensure the electrical dynamics. It has been shown in the literature that electrical dynamics and load variations accelerate the fuel cell degradation [6,7]. The objective of a hybrid electric architecture is to smartly split the power between the fuel cell and the battery. The fuel cell current is set up by the DC converter to minimize load variations and current dynamics while the battery provides the dynamic power solicitations to the load. Also, when the load demands are low, for instance when a vehicle is idled, then the fuel cell will recharge the battery pack.

1.3 Degradation mechanisms

1.3.1 Fuel cell stack

A PEMFC cell is mainly composed of bipolar plates, gas diffusion layers, two electrodes: anode and cathode, and a membrane between both electrodes. A fuel cell stack refers to the connection of cell in series. The fuel is supplied to the anode and the oxidant is supplied to the cathode. The membrane allows the passing of hydrogen ions and block the passing of electrons. The operating principle is simple: hydrogen ions flow through the membrane to the anode side and electrons flow through an external circuit creating an electric current. For years, many studies have shown that fuel cells degrade over time and depending on operating conditions [11]. Fuel cell degradation refers to an irreversible loss of power that can be supplied by the stack at a given operating point. The State of Health can be defined as the loss of power that the fuel cell can deliver at nominal operating conditions compared to a nominal reference at the Beginning of Life (BoL) [12]. Degradation mechanisms can occur at all components of a fuel cell stack : Bipolar plates, gas diffusion layers, electrodes and membranes [13] (cf. Table 1).

Table 1: fuel cell major degradation mechanisms

Component	Role	Degradation mechanisms	Causes
Membrane	Allow the protons travelling from anode to cathode; Block the travel of electrons; Participate to anode humidification;	Mechanical degradation (cracks and pinholes);	Non-uniform pressure; Drying; Thermal stress/cycling;
		Chemical degradation: poisoning;	Air impurities and fuel poisoning; Sealing; Peroxide attacks;
Bipolar plate	Isolating individual cells; Conduct the current between cells; Help in water and thermal management; Maintain mechanically the MEA; ensure good contact resistance;	loss of conductivity;	Corrosion;
		Facture and deformation;	Mechanical stress and Thermal stress/cycling;
Electrodes = GDL + CL	Electrical conductor used for electrons travelling from anode to cathode;	Losses of activation and conductivity;	Corrosion; Delamination; Oxidation; Flooding; Fuel poisoning;
		Decrease of reactant diffusion;	Mechanical stress;
Gas diffusion layers (GDL)	Ensure the gas diffusion from the flow fields to the active sites; Evacuate water;	Loss of hydrophobicity; Mechanical degradation;	Corrosion; Mechanical and thermal stress;
Catalyst layer (CL)	Assist the reactions of hydrogen oxidation and oxygen reduction;	Loss of conductivity; Loss of catalyst;	Corrosion; Mechanical and thermal stress;

As introduced in the table 1 above, not only the operating time leads to a loss of performance but also the operating conditions if they are improper. Indeed, a poor water management can lead to membrane dry out which increases resistive surfaces and can create cracks and pinholes or to cell flooding which increases mass transport losses. A bad air filtering can lead to a poisoning by contaminants and impurities [14], while a bad thermal management and Sub-zero temperature can lead to severe mechanical degradation. However, a loss of performance does not systematically mean a degradation as it can be reversible, depending on operating conditions [10,15,16]. For instance, humidity variations will directly affect the ohmic resistance value without it being irreversible [10].

Integrated in a vehicle, fuel cell systems are subject to rough operating conditions [4,5] such as dynamic load variations [6][7], air impurities, temperature and humidity variations, cold ambient temperature [8] and vibrations and shocks [9] which can accelerate the degradation of the system components. Developing reliable diagnostic and prognostic algorithms is then a crucial step to assist energy management strategies, ensure optimal operating conditions and predict the remaining useful life (RUL) of the system.

1.3.2 Batteries

The chemical reactions occurring within a battery cell are quite similar to the ones occurring in a fuel cell. The reactants of the electrochemical reactions in a battery cell are materials of anode and cathode, both of which are compounds containing lithium atoms. During discharge, an oxidation reaction at the anode produces positively charged lithium ions and negatively charged electrons. Then, the lithium ions travel through the separator (membrane) and the electrolyte and the electrons travel through an external circuit. They finally recombine at the cathode together with the cathode material in a reduction reaction. During charging, the roles of cathode and anode are reversed at the level of the negative and positive electrodes.

Like fuel cell, degradation mechanisms occur within a battery cell [17–20]. Schlasza et al. [19] proposed a detailed illustration (cf. Figure 2) highlighting all the degradation mechanisms occurring within a Li-ion battery cell.

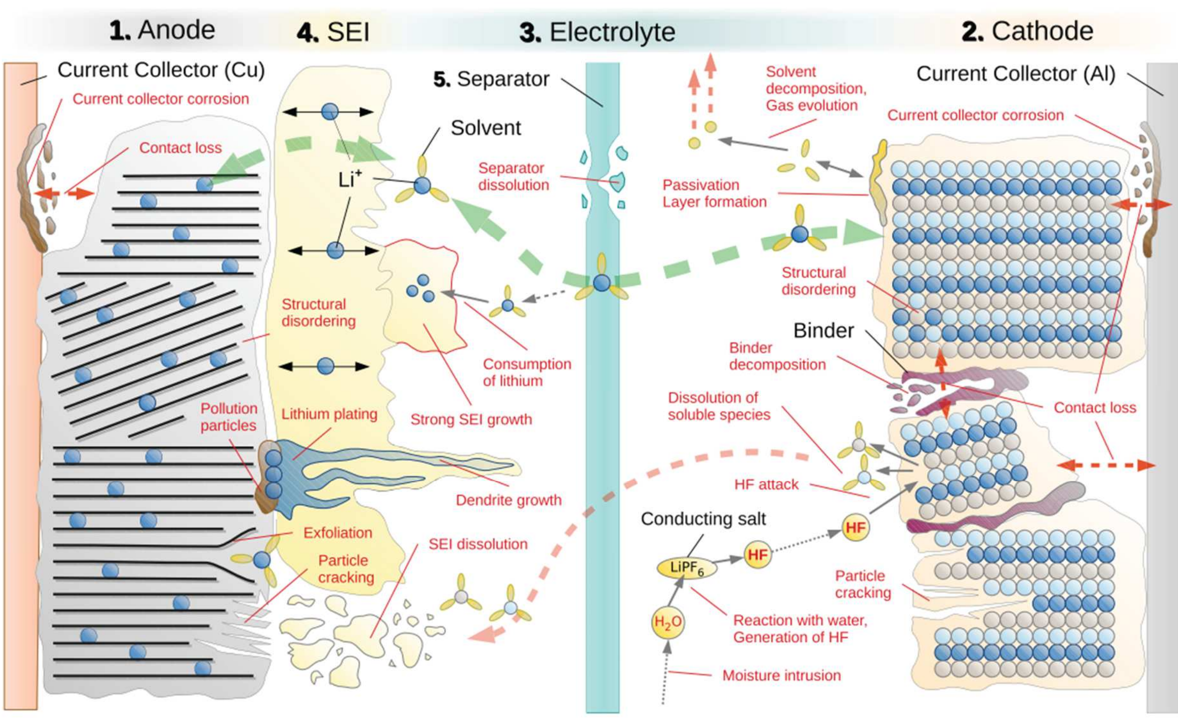

Figure 2: Main degradation mechanisms within a Li-ion battery cell [19]

Degradation mechanisms occur at all components level of the battery cell and they also lead to performance losses [17–20]. Table 2 classifies the major degradation mechanisms occurring within a battery according to their causes and consequences. In the case of batteries, performance loss can be distinguished in two terms: power fade and capacity fade.

- The battery capacity refers to the total quantity of electric charge that the battery can deliver. The capacity fade is usually defined as the ratio between the real available capacity and the available capacity at the beginning of life under the same operating conditions. The available capacity is also depending on the battery temperature and the current load.
- The power fade refers to the internal impedance increment which leads to a decrease of the available power that the battery can deliver. The available power is also depending on the temperature, the current rate, and the battery state of charge.

Table 2: Summary of main battery degradation mechanisms and consequences on performance degradation

Components	Degradation mechanism	Causes	Consequences on performance
Anode (including current collectors)	Lithium plating	Low temperature; Mechanical stress; Low voltage; Low SOC;	Capacity fade; Power fade;
	Morphological changes; particle cracking; structural disordering;	Cycling; Mechanical stress; High current	Capacity fade; Power fade;
	Current collector corrosion	Low voltage; Low SOC	Capacity fade; Power fade;
Cathode (including current collectors)	Current collector corrosion	Low voltage; Low SOC	Capacity fade; Power fade;
	Binder decomposition	High temperature; High voltage; High SOC;	Capacity fade; Power fade;
	Dissolution of soluble species	High Temperature; High SOC; High Voltage;	Capacity fade;
	Morphological changes; particle cracking;	Cycling; Mechanical stress	Capacity fade; Power fade;
Electrolyte	Electrolyte decomposition	High temperature; High voltage; High SOC;	Capacity fade
Surface Electrolyte Interface layer	SEI growth	Time; high temperature; High voltage; High SOC; Cycling; High current	Capacity fade
	SEI decomposition	high temperature; High voltage; High SOC; Cycling; High current	Capacity fade
Separator	Closing of separator pores	High temperature	Capacity fade; Power fade;

To summarize, like fuel cells, battery performances are affected by degradation mechanisms (cf. Table 2) and the operating conditions [16,21]. Integrated in a vehicle, batteries are subjected to high current dynamics and variable operating conditions. Then, battery capacity and internal impedance will vary all along the battery lifetime. A good estimation and prediction of the battery state of health (capacity and impedance) is thus required to guaranty good information about the available autonomy of the vehicle and the power that can be delivered.

1.3.3 Balance of Plant

Pukurshpan was one of the first authors to focus on the complete fuel cell system and consider the balance of plant besides the stack. He based his thesis works on fuel cell system modelling [22] and focused on stack and BoP performance modelling: namely air compressor, humidifier, air cooler etc. All the proposed models are deeply detailed and well explained but degradation phenomena were not considered in his study. Even nowadays, to the better of our knowledge, almost no paper has been published in the literature dealing with balance of plant (BoP) degradation. Most of studies dealing with BoP focus rather on fault conditions and detections at level of power converters [23] and hydrogen storage tanks [24].

1.3.3.1 Air supply line

Part of the air supply line, the air compressor is used to control the air flow rate as an input of the fuel cell cathode and the output air pressure of the stack. To prevent dehydration of the fuel cell membrane, the air flow is humidified and adjusted by the injection of water. This optimal air humidification is ensured by the humidifier. Obtaining optimal membrane water content is a key point when searching for better fuel cell performance and durability [10]. Some degradation mechanisms have been shown during the European project GiantLeap at the level of the air supply line: compressor and humidifier [25,26] (*https://giantleap.eu/*).

Table 3: Air supply line degradation mechanisms [25,26]

Component	Degradation mechanism	Causes	Consequences
Humidifier	Fouling;	Intake air impurities (particles, soiling, oil);	Fluids contamination; Fuel stack poisoning;
	Corrosion;	Intake air impurities (particles, soiling, oil);	Fluids contamination; Fuel stack poisoning;
	Fatigue; loss of performance;	Vibrations and shocks; Thermal stress; Aging;	Insufficient humidification; Stack drying;

Air compressor	Fouling;	Intake air impurities (particles, soiling, oil);	Fluids contamination; Fuel stack poisoning;
	Corrosion;	Intake air impurities (particles, soiling, oil);	Fluids contamination; Fuel stack poisoning;
	Fatigue; loss of performance;	Vibrations and shocks; Thermal stress; insufficient lubrification; Aging;	Increase of electric consumption;

1.3.3.2 Cooling system

The cooling system is mainly composed of a pump to make circulate the coolant fluid, a heat exchanger, a pre-heater, particles filters, ion particles filters, expansion tank, etc. A suitable cooling flow field designed with proper performance is of particular importance to increase the lifetime of a fuel cell [27]. Indeed, non-uniformity of temperature would cause the rate of electrochemical reaction to be variable at different places which can lead to hot spot formation and results in a stability and durability reduction. If the importance of a good thermal management has been proven in the literature, no degradation mechanism has been really reported. Logically, a fault condition in the cooling system would lead to over temperature in the fuel cell stack which can cause severe damages to the membrane that could be detrimental to the stack [28,29]. Also, a failure at the level of particles filters would lead to a too high conductivity of the coolant.

1.3.3.3 H2 line supply

The H2 line is composed of tank to store the gas, a pressure regulator, a pump to recirculate the gas, a purging valve, etc. Failure modes at level of hydrogen tanks have been investigated in the scientific literature. For type III composite hydrogen storage tanks, the most investigated failure modes are the burst, fiber damage and fatigue life [30]. Concerning the Type IV, the most investigated failure modes are the collapse and blistering of the liner, burst and damage [24]. Containers type IV are a widely used technology for high pressure hydrogen applications. It consists of an internal polymer liner acting as a diffusion barrier, a fibre reinforced composite and a metallic ending for pipe connections [31]. The collapse of internal liner has been observed especially at high rate decompressions [32].

1.3.3.4 Power Converter

In fuel cell vehicles, the main DC/DC converter between the fuel cell and the DC bus is a key issue [33]. If, no degradation mechanisms have been reported in the literature, power converters in fuel cell systems have been reported to be one of the most important failure sources. More precisely, failure sources are at level of power switches which are considered as the most delicate components in DC/DC converters [34,35]. This is why power switch fault diagnostic algorithms have been developed in order to guarantee systems' reliability and availability [23,35,36]. Otherwise, high and low frequency current ripples can affect fuel cell performance and also can lead to long-term degradation phenomena and then reduce the PEMFC lifetime [37–41]. Wahdame et al. [42] proposed a study which focuses on the influence of DC converter high frequency current ripples on the fuel cell degradation. For this purpose, two durability tests were carried out during 1000 hours on two PEMFC. During the first experiment which refers as a reference, the stack operated at 600W under stationary solicitations at roughly nominal conditions. The second experiment used a similar stack but operating under dynamic current solicitations. Results highlight a slight effect of high frequency current ripple on the fuel cell performance. According to [43], with a large current ripple, the operating point of the fuel cell must be chosen far from the most efficient point to avoid excessive losses and overheating. In this framework, new DC/DC converter topologies, such as interleaved boost converters, have been developed as this topology can minimize the current ripples. However, the loss of one leg in case of power switch fault can cause a drastic increasing of the current ripple. Guilbert et al. [36,37] proposed algorithms and mitigation strategies to overcome this issue. More details will be provided in the section 4. Thounthong et al. [40] presented a parallel converter modules with interleaving technique dedicated to a fuel cell. By this converter, fuel cell current ripples are reduced. Experimental results show excellent performance of the global system.

Whether it is a prognostic, diagnostic, or energy management algorithm, they are all based on system performance. As the performance is depending on operating time and on operating conditions, reliable algorithms must isolate and differentiate the causes of performance loss. For that purpose, degradation models are required.

1.4 System degradation modeling purpose

The purpose of a degradation model is to reproduce the dynamic behavior of the considered system according to the operating time and the operating conditions. Consequently, models must consider degradation and performance evolutions due to operating conditions variations otherwise diagnostic and prognostic algorithms will not be able to differentiate between abnormal operating conditions and degradation. The purpose of a degradation model is highlighted in the Figure 3. The objective is to reproduce the dynamic behavior, for instance output voltage, as a function of all the defined inputs such as current, temperature, operating time, pressures, humidity, etc.

A model can be used for diagnostic, prognostic, or energy management purpose.

It is worth noting that the tool to identify, study parameters or predict their evolutions is independent from the degradation model.

In this paper, a difference is made between the global prognostic/diagnostic approach and the model that governs the behavior (performance and degradation) of the considered system.

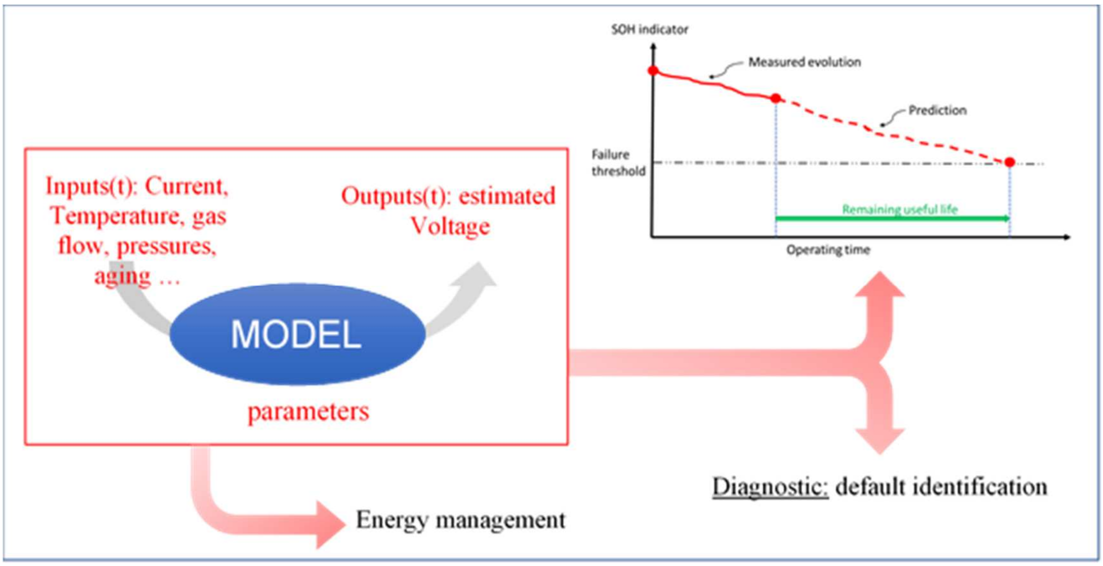

Figure 3: Purpose of fuel cell modeling

1.5 Classification of modeling methods

During the last decade, number of studies have focused on fuel cell and battery degradation mechanisms and behavior modeling. However, studies that focused on modeling the degradation laws are much less numerous. Fuel cell and battery systems modeling methods can be classified according to three different categories: Physics-based models, data-driven models and hybrid models (cf. Figure 4).

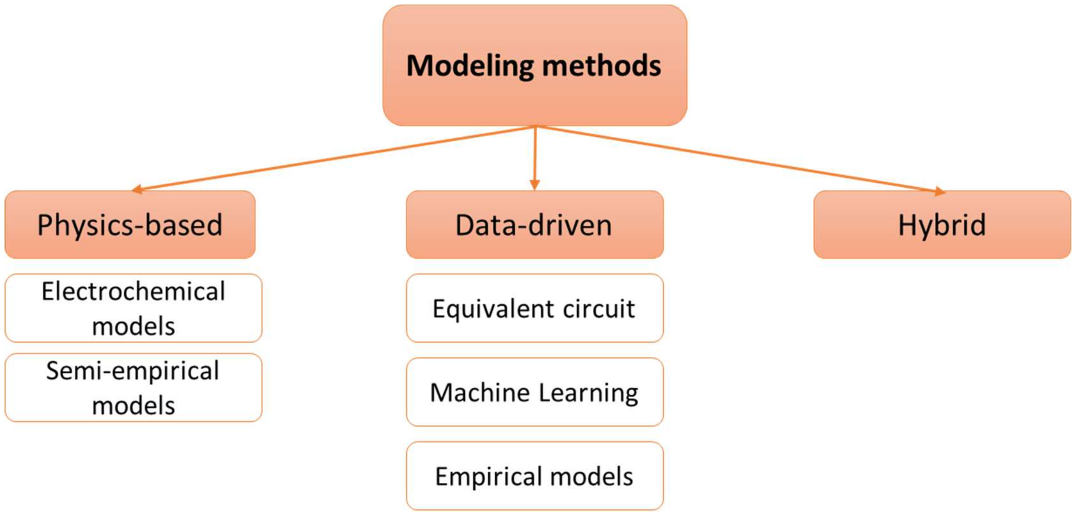

Figure 4: classification of PEMFC modeling methods

Those three categories can be defined as follows and will be more detailed. Definitions and reference will be given, in the next sections.

- **Physics-based models:** they can be seen as white or grey box models. They are based on the physical equations of the considered system. They can be mechanistic (white models), semi-mechanistic or semi-empirical (grey box models). It is worth noting that semi-empirical models are mainly based on physical equations of the considered system but can integrate one empirical variable like a resistive parameter for instance.
- **Data-driven models:** they can be seen as black box models. They are based on the measured data used to learn the behavior of the considered system and build a model. Empirical models, electrical circuit models or neural network models are considered as data-driven models.
- **Hybrid models:** they combine both physics-based models and data-driven models.

The contribution of this paper is to provide a comprehensive and critical review of the existing methods to model fuel cell stack, balance of plant and battery, considering the degradation of system performance. At first, the paper will focus on the fuel cell stack modeling methods. Then, the focus will be on the balance of plant starting with the battery, followed by the fuel cell ancillaries.

2 Fuel cell existing degradation modeling methods

2.1 Physics-based models

Physical models can be mechanistic, semi-mechanistic and semi-empirical and are based on system physical equations to reproduce its behavior considering the degradation. The main advantage is that physical models do not require a large amount of data then they present a good generalization ability. Moreover, as they are based on system internal physical equations, users can observe and correlate the changes and the evolution of internal physical states and parameters of the system. Consequently, physical models require a good understanding of the system degradation laws but also good knowledge about internal parameters. This can be a major disadvantage as PEMFC degradation mechanisms and laws are not fully understood yet and most of the time information about internal system parameters are not given by manufacturers due to confidentiality issues.

Zhang et Pisu. [44] proposed a physical fuel cell stack degradation model with the Electro-Chemical Active Surface Area (ECSA) as an aging parameter of the fuel cell degradation process. Based on an Unscented Kalman Filter (UKF), the proposed algorithm is able to estimate and predict the ECSA degradation all along the stack operation. The proposed method was successfully applied in simulation with dynamic solicitations. However, the only aging parameter, the ECSA cannot fully model the degradation. Indeed, the ECSA reduction is not the only degradation phenomenon in a fuel cell stack then other aging parameters should be considered.

Zhang et al.[45] proposed a degradation model used for prognostic purpose. The proposed method combines the information of two different sources. The first one receives a signal directly observable and related to the stack voltage. The second one is fed periodically by measurements from physical characterizations of the stack. Characterizations give good information about the SOH evolution. Then, the prognostic procedure is implemented using Particle Filtering (PF). The fuel cell model is a quasi-static model based on the polarization curve (cf. equation 10). The degradation is considered through one variable γ which is associated to the internal resistance r and the open circuit voltage E (cf. equation 11). Finally, y is predicted by a particle filter.

$$V_{stack} = n(E - r.j - A.ln\left(\frac{j}{j_0}\right) - m_1.exp(m_2.j)) \qquad (10)$$

$$r(t) = r_0(1 + \gamma(t)), E(t) = E_0(1 - \gamma(t)) \qquad (11)$$

Where r is the internal resistance, j is the operating current density, A is the Tafel coefficient, j_0 is the exchange current density, $m_1$et m_2 are the mass transfer constants, r_0 and E_0 are the initial values of r and E.

The proposed method shows promising results. In this case, only one degradation coefficient is considered which might not be sufficient to fully reflect the performances losses. Bressel et al. [46] proposed a similar work based on the use of a physical model to estimate the state of health of a fuel cell stack and to predict its degradation evolution. One aging parameters α is integrated in the ohmic losses and diffusion losses as in the following equations (cf. equation (12),(13)).

Authors stated that for a given current, it is not possible to estimate separately the influence of parameters on a cell voltage [46]. For this reason, they made the choice to link the change in the stack resistivity and the current limit with the single parameter α. Then, instead of a particle filter, in this study, a Kalman filter is used to estimate and update the aging parameter α and its derivative β which represents the SOH evolution.

$$V_{stack} = n(E_0 - Ri - ATln\left(\frac{i}{i_0}\right) - BTln\left(1 - \frac{i}{i_L}\right)) \quad (12)$$

i the stack current, n the number of cells, T the temperature, A the Tafel constant, B a concentration constant.

$$\text{With: } R = R_0(1+\alpha), i_L = i_{L_0}(1-\alpha) \quad (13)$$

With R_0 the beginning of life ohmic resistance at nominal operating point. Like the previous study ([45]), one degradation coefficient is considered and associated to ohmic and diffusion losses by making the hypothesis that they are mirrored. To improve this model and better reflect degradation laws, more degradation coefficients could be integrated to consider separately activation, ohmic and diffusion losses.

Zhou et al. [47] proposed a degradation model for PEMFC stack performance based on a multi-physics aging model and a particle filter. The proposed model is based on physical fuel cell equations. To consider performance degradation, three aging coefficients (α, β, γ) are integrated in major internal physical aging phenomena equations: ohmic losses, reaction activity losses and reactants mass transfer losses. At first, the aging parameters are initialized by fitting the polarization curve at the beginning of life. The prediction is governed by two phases: learning phase and prediction phase. For the learning phase, a particle filter framework is performed to study the aging behavior and to update the aging parameters. Then, the suitable fitting curve functions are identified to satisfy the aging parameter' evolutions and are used to extrapolate and predict their evolution during the prediction phase to finally predict voltage evolution.

Although this method shows satisfying results, its accuracy depends on the operating conditions variations. Indeed, the proposed prediction method is based on the identification of the good fitting curves, then if the operating conditions change, the fitting functions must be changed. Therefore, the proposed modeling method would rather be dedicated to steady state operations and constant operating temperature.

Jouin et al. [48] proposed a prognostic tool based on a similar model. However, they consider more aging parameters, eight precisely: α_a and α_c which are the charge transfer coefficients at the electrodes, i_{loss} which refers to the hydrogen crossover current, $R_{ion}, R_{elec}, R_{cr}$ which are respectively the ionic, the electronic and the contact resistances, B_c which is an empirical parameter used to consider the effect of water and gas accumulations and $i_{max,c}$ which is the limiting current at the cathode. Coefficients are initialized in a first step empirically based on literature and initial polarization curve. In a next step, parameters are updated with a particle filter based along a long-term durability experimentation. Results are promising but as well as the others, the presented models are limited to a quasi-static behavior which may be not sufficient to fully reproduce the dynamic behavior of a fuel cell integrated in transportation application.

Synthesis, Physics-based models:

To summarize the literature survey about physics-based models:

Advantages:

+ Can be easily implemented and performed online.
+ Can give information about the internal physical parameters.
+ Can be used to investigate physical phenomena occurring within a fuel cell.
+ Good generalization ability.

Limitations:

- Generally static or quasi-static models. Transient phenomena are not considered.
- Require a good understanding of the system behavior and its degradation laws.
- Require knowledge about internal system characteristics.

These methods have the advantage to be easily performed online as they are based on physical equations that govern the system then they do not require a large amount of data. That is the reason why they provide a good generalization ability. Moreover, as they are based on physical equations, they can give information about the internal physical parameters of the system. This information can be used in the future for diagnostic or prognostic purpose. However, they are generally limited to static or quasi-static equations and transient phenomena are not considered. Moreover, as they are based on system physical equations, they require knowledge about internal system information and a good understanding of the system behavior and degradation laws.

2.2 Data-driven models

The data-driven models are typically empirical, statistical, or mathematical models and rely on experimentations and data analysis. Generally, they are based on the use of equivalent circuit models or machine learning tools to build black box models. A major advantage is that these methods do not require knowledge about internal system parameter information. Moreover, these methods do not require an in-depth understanding of the system degradation laws. However, this can also be a problem as users cannot observe and correlate the change of internal state parameters of PEMFC system. This can lead to a lack of important information for diagnostic and decision-making purposes. Another major disadvantage is that these methods need a large amount of representative training data to build the model and as the degradation laws differ according to the system, the aging data must come from systems of the same type and manufacturer. Consequently, the generalization ability of such models is rather poor.

2.2.1 Artificial neural networks

Artificial neural networks have been widely used for prognostic purpose in order to model the performance evolution of a fuel cell stack [49]. Several structures of neural network have been presented such as feed forward neural network (FNN), recurrent neural network (RNN), long short term memory neural network (LSTM), echo state network (ESN) [50,51]. Neural networks are part of artificial intelligence as they need a training phase to learn the dynamic behavior of the considered system according to the defined inputs and reproduce it.

2.2.1.1 Feed forward neural networks

Napoli et al. [52] used a classical neural network to model stack voltage evolution and cathode temperature of a 5kW fuel cell. Results showed that the proposed method can reproduce the impact of different stoichiometric ratio on the voltage under different operating conditions. Chen et al. [53–55] used neural networks in three studies to model the stack voltage evolution of a fuel cell integrated in a lightweight postal vehicle. Although the proposed methods seem interesting, the fuel cell operated for 50 hours only, which allows hardly to analyze the degradation. Moreover, the model is built with a learning phase performed on the 40 first operating hours, which seems to be not enough to describe the fuel cell performance degradation. Indeed, it has been shown in the literature that nowadays, fuel cell durability considering good operating conditions can be over hundreds of hours [10,12,48,51]. Besides, the validation of the model is performed with the data measured during the 10 last hours, which is not really significative to verify the proposed model considering durability of hundreds of hours.

2.2.1.2 Recurrent Neural networks

Recurrent neural networks are preferred to model nonlinear dynamic temporal signal as the output at the last step is injected as an input at the present step (cf. Figure 5). Thanks to that, signals whose evolution at present step depends on the last steps can be considered for that type of NN.

Liu et al. [56] proposed a PEMFC degradation modeling method based on the use of Long Short-term memory (LSTM) recurrent neural network (RNN) to estimate the remaining useful life of a PEMFC. Results are verified with experimental aging data and show that the prediction accuracy of the proposed method is 99.23% and the root mean square error is 0.003. Ma et al. [57] also proposed a method based on the use of LSTM. The method was verified experimentally, and results show promising accuracy. LSTM neural networks are powerful tools to build nonlinear data-based model. However, computational requirements can be very expensive and time to build the model during the learning phase can be consequent for large databases.

2.2.1.3 Reservoir computing

Jaeger et al. introduced Echo state network (ESN) in the early 2000s [58]. This architecture of recurrent neural network reproduces more faithfully the functioning of the human brain as the hidden layers are replaced by a reservoir of neurons (cf. Figure 5). The input weight and reservoir weight matrices are both created randomly. Only the output weights are optimized by a multi linear regression then computational requirements are much lower compared to feedforward neural network and LSTM neural networks.

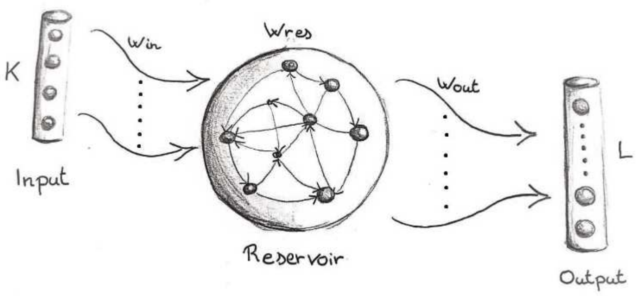

Figure 5: ESN architecture [12]

with:

- N_{res} the number of neurons in the reservoir.
- K the number of inputs.
- L the number of outputs.
- W_{in} the input weight matrix.
- W_{out} the output weight matrix.
- W_{res} the reservoir weight matrix.

Morando et al. [51] used an echo state network to model voltage evolution of a fuel cell stack and forecast its end of life. The proposed study highlights that the computational requirements of reservoir computing are advantageous compared to classic recurrent neural networks. The authors forecast the voltage evolution over 1000 hours with a mean average percentage error lower than 5%. Hua et al. [59] also proposed a degradation fuel cell stack model based on the use of ESN. They also assure that the specific ESN architecture leads to a better performance especially in reducing the computational costs. The proposed study compares two ESN structures. The first one is a single input structure and the second one is a multiple-input structure. Results showed that the multiple-input structure leads to better performance under both steady state and dynamic operating conditions. Both studies show good prediction results however, the system operated under static current loads and constant temperature, which does not reflect the reality for automotive applications.

Vichard et al. [12] used an ESN to model a complete open cathode fuel cell system degradation operating under actual conditions. The purpose of the experimentation is to approach at best on a test bench, the operating conditions of a lightweight postal hydrogen vehicle. The fuel cell system is integrated in a range extender architecture and operates at nominal operating point (1kW, 50A) under variable ambient temperature. Results of the aging experimentation showed that in addition to the operating time, ambient temperature variations, due to open cathode configuration has an impact on the system performance degradation. To model this degradation, the authors preferred the use of an ESN as it does not need physical system information and due to the low computational requirements. As illustrated in the Figure 6, the ESN inputs are the ambient temperature, the operating time which is the image of the aging and the stack voltage at the last step.

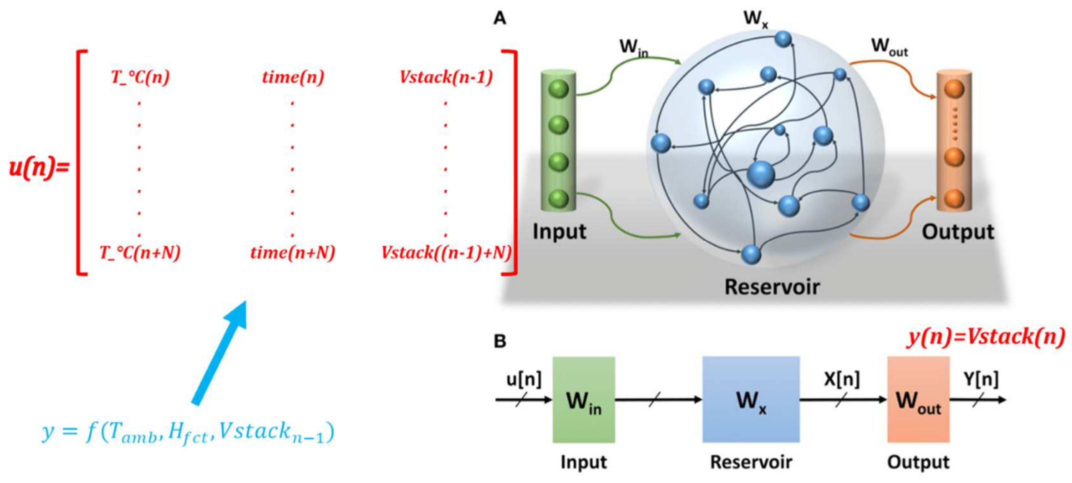

Figure 6: ESN architecture in [12]

The model shows a satisfying accuracy and is able to predict stack voltage evolution accurately over 2000 hours. Nevertheless, to achieve a good accuracy, the model must be built considering the entire ambient temperature range otherwise it is not able to reproduce completely the system dynamic behavior. Therefore, a consistent database is required to build a suitable model.

2.2.2 Equivalent circuit model

Electrochemical impedance spectroscopy can be used to build a fuel cell model [16,60–64]. As illustrated in the Figure 7 [16], the Nyquist diagram can be used to identify the parameters of an equivalent circuit model.

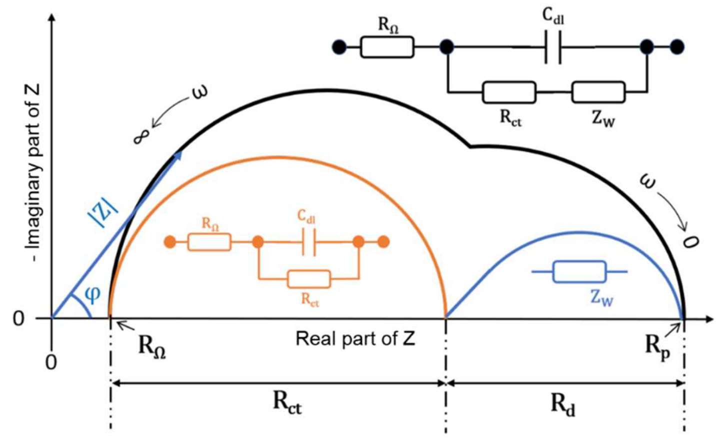

Figure 7: Modeling based on EIS concept [16]

Lee et al.[65] proposed a degradation modeling method to estimate remaining useful life of a single PEM fuel cell through an equivalent impedance model based on electrochemical impedance spectroscopy (EIS). The author uses charge transfer resistance and double layer capacitance as major parameters for judging degradation. Gaumont et al. [64] used the EIS to study the effect of humidification and degradation on the electrodes. Fouquet et al. [61] used the EIS to build a Randles equivalent model based on experimentations carried out under different operating conditions. The showed that a Randles model augmented with a CPE (Constant Phase Element) was found to be an accurate model of the fuel cell dynamic behavior over a wide range of operating conditions. Other studies used the EIS to estimate degradation by measuring ohmic and diffusion resistances [10,50,62,64,66]. Electrochemical impedance spectroscopy can also be efficient for diagnostic purpose by detecting abnormal operating conditions such as drying, flooding, poisoning etc. [10,60,61,67]

EIS is a very reliable tool to get information about the fuel cell degradation and dynamic behavior for modeling and diagnostic purpose. It is worth noting that EIS integration in embedded application needs to be well mastered. Indeed, measures strongly depend on operating and ambient conditions and they must be strictly identical to correlate the data. Therefore, EIS is rather dedicated to laboratory applications. However, some studies have shown that it is possible to implement EIS measurements on-board [68]. Depernet et al. [69] proposed a study dealing with integration of EIS functionality in PEMFC power converter. The fuel cell is electrically coupled to a DC bus using a DC/AC/DC converter based on an inverter stage, a high frequency power transformer stage and a rectifier stage. Impedance measure is performed thanks to the power converter without any additional hardware. However, they specified that during the impedance measurement, the operating point of the PEMFC needs to be maintained fixed. This means that the measurement must be performed during vehicle stops or the battery must be able to provide alone the current dynamics to the vehicle. Wang et al. [68] also proposed a study focusing on EIS on-board integration. The study proposes an on-board EIS detection strategy based on a fuel cell stack connected step-up converter. Like the previous study, no additional hardware is required. Simulations carried out under different operating conditions showed that the proposed strategy is able to reproduce Nyquist diagram faithfully and detect fault conditions. However, according to the impedance spectroscopy principle, the operating point has to remain strictly identical in order to correlate the data [69].

Synthesis, data-driven models:

To summarize the literature survey about data-driven models:

Advantages:

+ Does not require knowledge about internal parameters of the system.
+ Does not require an in-deep understanding of system behavior and degradation mechanisms…

Limitations:

- .. But cannot be used to investigate physical phenomena occurring within a fuel cell.
- Can require a large amount of data to build an efficient model.
- Require exhaustive database for the model be reliable.
- Poor generalization ability.
- Poor extrapolation ability.

Data-driven models do not require knowledge about the internal parameters of the system which is a real advantage when the knowledge about internal parameters of the studied system is limited. Moreover, they do not require an in-deep understanding of the system behavior and the degradation laws as the model parameters are identified empirically based on a database. This advantage can also be a limitation as a large amount of exhaustive data is required to build an efficient model. As they are based on data, data-driven models possess poor generalization and extrapolation ability.

2.3 Hybrid methods

The purpose of hybrid models is to combine a physical based model with a data-driven model in order to cumulate advantages of both methods.

It is worth noting that a model is a tool who reproduces the system behavior, then the algorithm performed to identify parameters or predict their evolutions is independent from the model. In this paper a difference is made between the global prognostic approach and the model that governs the behavior of the considered system. Then these published models are considered here as being physical based and not hybrid. A hybrid model combines a physical based model and a black box model to reproduce the dynamic behavior of the considered system.

Lan et al. [70] proposed a hybrid model of a fuel cell stack. They proposed a model that covers the multi-physics transients involved in the flows of electric current, hydrogen and other gases and the enthalpy. First, the methodology to model mutually coupled electric, pneumatic, and thermal transients by coupling the use of physical based model and electrical circuit models, was applied to model a PEM fuel cell. The overall multi-physics model is then validated with experimental data.

Lechartier et al. [71] proposed a degradation model of a fuel cell stack composed of a static part and a dynamic part that are independent. The static part is based on equations describing physical phenomena. The dynamic part is based on an electrical equivalence of the physical phenomena. Parameters of the models are updated based on characterizations such as polarization curves (IV) and electrochemical impedance spectroscopy (EIS). Although the accuracy is promising, the proposed method shows limitations. Indeed, models are based on periodically characterizations, polarization curves and EIS, which can be hard to perform on-board for transportation applications. Zhong et al. [72] proposed an approach to model a PEMFC at system level based on the development of a hybrid model. As a hybrid approach, this model is composed of two parts. The first part is an empirical model based on the use of least-square support vector machine to consider current and temperature. Parameters of the model are identified through a PSO optimization algorithm and results were verified experimentally. The second part is a pressure-incremental physics-based model to consider cathode and anode pressures evolution. The proposed approach is able to reproduce polarization curves considering temperature and pressures variations. However, the proposed model should be used under limited pressure variations, as too important variations will affect fuel cell voltage and current too significantly. Therefore, the fuel cell will move away from nominal reference conditions under which the proposed pressure-model was calibrated.

Synthesis, hybrid models:

To summarize the literature survey about hybrid models:

Advantages:

+ Theoretically, can combine advantages of both physics-based and data-driven approaches...

Limitations:

- …But then, can also combines drawbacks.
- Nowadays, very limited number of published PEMFC hybrid models.

The purpose of hybrid models is to combine at best the advantages of both physical based and data-driven models. The limitation is that drawbacks can also be combined. For this reason, a good compromise has to be identified to keep at most the advantages while reducing at best the drawbacks. Thanks to a hybrid model, the behavior at the beginning of life (no degradation) could be reproduced without the need of a large amount of data and, in parallel, a machine learning tool can be performed online to learn and predict performance losses all along the system lifetime. However, nowadays there is a very limited number of published PEMFC hybrid models, but such models seem to be a very good modelling method to be focused on in the future.

3 Batteries

As mentioned previously, battery performances tend to decrease overtime and according to operating conditions: internal impedance increment and loss of available capacity. Internal impedance directly refers to the maximal power that the battery can deliver, and available capacity directly refers to the maximal quantity of electrical charges that the battery can deliver. For numerous applications, these two characteristics can be of particular importance. Integrated in a hybrid fuel cell system, the battery capacity and its internal impedance will be directly linked to the energy management strategy and will affect the operation of the fuel cell [73]. For these reasons, degradation models and prognostic approaches need to be developed. In the case of batteries as well, models can be distinguished according to this classification: physics-based models, data-driven models, and hybrid models. In practice, battery state of health indicators, i.e., internal impedance and capacity, cannot be measured directly. Consequently, degradation models must be able to consider, estimate and reproduce these indicators all along the battery lifetime.

3.1.1 Physics-based models

Electrochemical models are usually used to understand electrochemical phenomena [74] or to study a specific phenomenon such as phase change, effect of the porous structure of the electrodes or again relaxation phases [75]. Moura et al. [76] proposed to use an SEI film formation model to express the degradation degree of a battery cell. This model integrates a large number of state variables and a large set of nonlinear algebraic constraints, which leads to high computational cost requirements. Moreover, due to the complexity of this model, it can be hard to implement it on controllers. Xia et al. [77] proposed a multiphysical model of a Li-ion battery. The proposed model couples electrochemical, thermal and SEI formation modeling considering fluid and electrical dynamics. A life model including capacity fade and percentile life was also presented. Experiments have been carried out on test bench to verify the proposed methods. Results are very promising and show that the degradation rate of battery first decreases and then accelerates all along the battery lifetime. This model could be used for in-depth analyze of physical phenomena occurring within a battery cell and to follow parameters evolution in order to better understand degradation laws. However, although this method proposes promising results, the model was verified only with charge and discharge cycles which do not fully reflect actual operating conditions. Moreover, due to the complexity of the proposed method, it is rather dedicated for laboratory and research uses. Other studies have focused their works on the use of electrochemical physics-based models [78–80].

Synthesis, Physics-based models:

To summarize the literature survey about battery physics-based models:

Advantages:

+ Can be used to understand electrochemical phenomena.
+ Can be used for in-depth analyze of physical phenomena occurring within a battery cell.

Limitations:

- Many physico-chemical parameters are required: can be complex and hard to be identified.

Physical and electrochemical models can be used to study and understand the electrochemical phenomena occurring within the battery cells. Physics based electrochemical models can be complex to developed and to implement as they need many physico-chemical tuning parameters and thus require important computational cost requirements. For these reasons, battery physics-based models still remain rather dedicated for laboratory uses.

3.1.2 Data-driven models

3.1.2.1 Equivalent circuit models

Equivalent circuit model (ECM) have been widely used to model batteries and estimate their state of health [21,81,82]. These methods present low complexity, low computational requirement, and acceptable accuracy. As highlighted on Figure 8, ECM integrates resistive and capacitive elements. By adding R//C circuits, the model can reproduce the dynamic voltage response of a battery cell which is a sum of transient signals of different durations. R//C circuits are used to represent impedances linked to the charge transfer effects, double layer effects, diffusion, and relaxation phenomena [83,84]. The number of R//C circuits depends on the desired accuracy and computing requirement limitations. Structure can be found considering only one R//C circuit [85–88], two R//C circuits [82,89–91], or three R//C circuits [92–94].

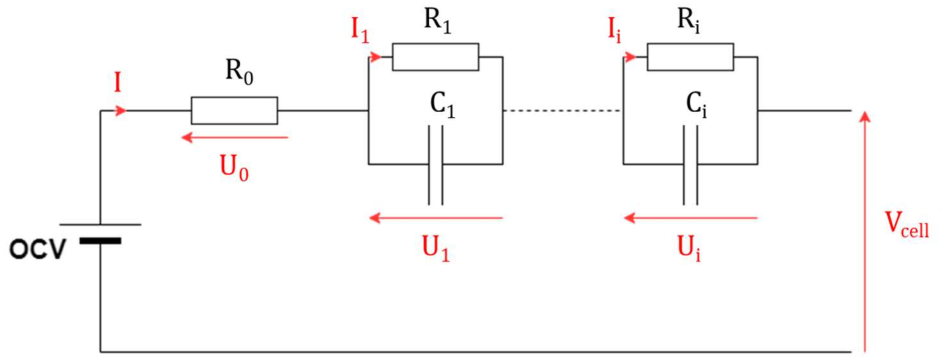

Figure 8: Battery equivalent circuit model

This kind of model can be used to reproduce the dynamic voltage response of a battery cell but also to estimate the state of charge and both health indicators: the internal resistance and the capacity [21].

Various methods can be used to identify the parameters of the ECM. Ren et al. [95] proposed a method based on the use of an ECM coupled to a Kalman filter to identify model parameters and estimate the state of charge, and both health indicators: internal resistance and capacity of a battery pack. The proposed method has been verified experimentally thanks to Hybrid Pulse Power Characterization (HPPC) profiles and showed promising results. Shen et al. [89]

proposed a method to estimate concurrently the state of charge and the state of health of a li-ion battery integrated in real-time applications. The battery capacity is estimated with respect to the accumulated electric charge between two specific moments with online OCV identification. Based on the capacity update from SOH estimation, the state of charge can be estimated depending on the battery aging. Results are promising but this method shows some limitations. Indeed, it is based on OCV curves then it can be very hard to be applied on technologies such as LiFePO4 where OCV curves are majorly flat. Several studies proposed methods based on the use of an ECM coupled with Kalman filters to estimate the state of charge, and health indicators [96–100]. Remmlinger et al. [101] also proposed a method based on the use of a Recursive Least Square algorithm (RLS) to identify the parameters of a two R//C circuits ECM. Thanks to this model they can observe the internal resistance evolution as a function of the temperature and the aging all along the battery operating time. Vichard et al. [16] proposed a method based on the use of a three R//C circuits ECM. Firstly, the model parameters are identified with an optimization algorithm. OCV and resistive parameters are defined as a function of the temperature and the state of charge. The internal resistance is defined as the sum of the resistive elements of the ECM. The model is then verified thanks to real driving cycle and shows very promising accuracy. Secondly, an extended Kalman filter is used to estimate concurrently the state of charge and the battery capacity.

Synthesis, equivalent circuit models:

To summarize the literature survey about ECM:

Advantages:

+ Easy to implement.
+ Good accuracy
+ Can be used for SOC and SOH estimation

+ Can be used to estimate internal resistance.
+ Does not require full system knowledge…

Limitations:

- …But dependencies on operating conditions and aging must be translated mathematically.

ECM are often used for embedded applications as they are very easily to be developed and computed within cheap microcontrollers as they do not require high computational requirement. Moreover, they provide a good accuracy and can be used for battery SOC and SOH estimation or even to estimate the internal resistance. As data-driven models they are not based on physical equations but on equivalent electrical parameters, thus such models do not require full system knowledge. However, dependencies linked to operating conditions and battery aging must be translated mathematically which requires a minimum of knowledge about system dynamic behavior.

3.1.2.2 Empirical and Machine learning models

Data driven approaches have also been used to model battery, estimate the state of health impact and model degradation phenomena such as capacity fade. Redondo et al. [102] proposed an approach to model the capacity fade of an NMC battery cells considering both calendar and cycling aging. For that purpose, they carried out an accelerated aging experiment to investigate battery degradation at SOC levels and current solicitations representative of the actual operating conditions as integrated in a vehicle. Model's parameter identification is composed of two steps. In a first step, a calendar ageing model is proposed based on the Eyring law. Parameters are identified through a multi-linear regression with the use of the experiment data. In a second step, cycling aging data from experiments are combined to the calendar ageing model to identify the combined ageing model parameter through an optimization algorithm. Based on only two differential equations and seven parameters, the proposed model is able to reproduce the capacity evolution of a battery cell considering both cycling and calendar solicitations as integrated in a vehicle. The proposed approach is very promising and opens number of perspectives. However, the proposed approach does not consider temperature variations as it would be in an actual vehicle. Also, the proposed approach relies on test campaign to identify model's parameters which can be costly. Artificial neural networks have also been used to model performance degradation of batteries. Like for the fuel cell, these approaches do not consider physical laws of the battery cells as they are fully based on measured data and a learning phase. Eddahech et al. [103] proposed a method based on the use of a RNN to estimate the SOH and build a degradation model. A model is built thanks to EIS measurements performed at different aging levels and under different operating conditions. Then parameter evolutions are learnt by the RNN. Finally, the neural network degradation model is able to reproduce and predict the dynamic behavior considering health indicator evolution. Shen et al. [104] proposed a method based on the use of deep learning tools for cell-level capacity estimation based on the voltage, current and charge capacity measurements during partial charge cycles. Results showed that temperature variations can result in significant changes to the capacity estimation. Moreover, the method has not been verified with real application data. Electrochemical Impedance Spectroscopy (EIS) has also been used to identity parameters of a battery model [105]. Zenati et al. [106] and Schweiger et al. [107] proposed methods based on the use of the fuzzy logic coupled to EIS measurements to estimate and predict battery health indicators. The methods showed promising accuracy however, they are based on characterizations through EIS measurements. For these reasons, these methods are not well dedicated to on-board applications.

Synthesis, machine learning models:

To summarize the literature survey about battery machine learning models:

Advantages:

+ Does not require to link model parameters with aging and operating conditions.

Limitations:

- Require large amount of data to build the model.
- Require exhaustive database.
- Poor generalization ability and extrapolation ability.

Contrary to ECM, machine learning, and black box models do not require any knowledge about system internal parameters and dynamic behavior to be built. However, they require a large amount of exhaustive data to be reliable. For this reason, generalization and extrapolation abilities are poor. Moreover, machine learning, and black box models do not describe the internal mechanism, chemical reaction, physical behavior, and structure of the battery. For these reasons, these models are not suitable if an in-depth investigation on the mechanisms and reactions occurring in a battery cell is needed.

4 Balance of Plant

4.1 Air supply system

4.1.1 Air compressor

Some research works have been published regarding fuel cell air compressor, air supply and control strategies [108]. According to the Nernst equation, performance of fuel cell improves as the reactant gases pressure increase. Furthermore, a higher oxygen concentration, associated to possible high current delivery, will improve reaction kinetics, and increase water production, thus resulting in better humidification conditions. Moreover, integrated in a vehicle a fuel cell is subjected to load variations and start-stop conditions. These operating conditions are highly related to water management and gas transport which can significantly impact the system performance and durability [4,7,10]. Besides, higher gas pressures do not only improve fuel cell performance but also improves water management [108]. For these reasons, set up optimized air supply control and humidification strategies is of particular importance. These strategies are mostly based on compressor control oriented models [108,109].

4.1.1.1 Physics-based models

All studies who proposed a physics-based air compressor model used the model expressed in the following equations [110–114]. The compressor angular speed ω_{cp} is governed by the combined compressor motor inertia J_{cp} according to the expression (14).

$$J_{cp}\frac{d\omega_{cp}}{dt} = \tau_{cm} - \tau_{cp} \tag{14}$$

Where $\tau_m$et τ_{cp} denote the compressor accelerating torque and load torque, respectively.

$$\tau_{cm} = \frac{\eta_{cm}K_t}{R_{cm}}(\, \upsilon_{cp} - K_v\omega_{cp}) \tag{15}$$

$$\tau_{cp} = \frac{C_pT_{atm}}{\eta_{cp}\omega_{cp}}\left[\left(\frac{P_{sm}}{P_{atm}}\right)^{\frac{\gamma-1}{\gamma}} - 1\right]W_{cp} \tag{16}$$

Where η_{cm} is the motor efficiency, K_t is the sensitivity of torque, K_v is the constant of counter electromotive force and R_{cm} is the motor resistance, υ_{cm} is the control voltage of the compressor. C_p is the specific heat of air, γ is the specific heat capacity ratio, $P_{atm}$is the pressure of atmosphere, $P_{sm}$is the pressure in the supply manifold, and W_{cp} is the flow rate of the air from the compressor to the supply manifold.

4.1.1.2 Data-driven models

Some other research works proposed data-driven approaches to model air compressor. Hernandez et al. [115] published in 2005 a study dealing with modeling of a motor compressor group feeding a PEMFC by using recurrent neural network. The comparison between the simulation results and experimental data shows a very good accuracy of the proposed model. Zhao et al. [109] proposed an air compressor model based on the use of a back propagation neural network in order to reproduce the air mass flow a as function of the speed and the pressure. The model was built based on experimental data. Deng et al. [110] proposed a data-driven method based on the use of NARMAX modeling coupled with a recurrent neural network. The NARMAX model is an equivalent time-varying linear model and the time-varying parameters are identified by an RNN. Some others studies based their model on a ninth order model [116,117].

4.1.2 Humidification system

Coupled to the air compressor, humidifier is of particular importance in order to add water content to the input air without what fuel cell membranes could dry out. Some approaches to model fuel cell humidification process have been published during the last years. Adair et al. [118] proposed a quasi-static distributed parameter model of the water circuit of a typical cell system. They showed that the proposed modelling is able to replicate physical phenomena. Chen et al. [119] proposed a physics-based model for membrane humidifier integrated in an automotive fuel cell system. The proposed 4 states humidifier model describes the transient behaviour of the humidification process and captures the time varying effect of the flow rate, temperature, pressure, and relative humidity. Based on this model they proposed a proportional feedback control algorithm to maintain proper membrane humidification level within the fuel cell stack. Chen et al. [120] proposed another physics-based approach. This research works present at first an experimental study carried out on a membrane humidifier. In a second time they proposed a thermodynamic humidification model for a PEMFC humidity control. The proposed approach considers both steady-state and dynamics phenomena. Model is verified experimentally and show good accuracy. Nielsen et al. [121] proposed a steady-state distributed parameter model of a membrane humidifier. They showed that the proposed model is able to replicate the physical phenomena occurring during real life conditions in membrane humidifier. However, the proposed model does not include the influence of dynamic phenomena in the water diffusion process. Liu et al. [122] also proposed a physics-based model of the humidification process in the cathode of a PEMFC. For that purpose, they developed a control-oriented model of the humidifier air supply line to drive the dry inlet air to be humidified. Solsona et al. [123] proposed a physics-based control oriented fourth-order non-linear model of an air humidifier. They proposed a generic approach to the readers to be able to model similar systems whatever the scaling factors and the operating conditions. The model considers both heat and mass dynamics. The proposed method has been verified experimentally under both static and dynamic operating conditions and considering the full range of operation. Results show a good accuracy. Finally, the model was used as a base of designer for a non-linear control strategy. All these approaches are physics-based and rely on similar physical equation describing humidification process of a PEMFC.

All the models show promising accuracy considering both steady-state and dynamic operations. Such models provide an important tool for external fuel cell humidifier design, simulation and to develop membrane humidification control strategies. However, these approaches do not consider degradation phenomena and focus only on modelling behaviour and fluid exchanges. Several studies highlighting the impact of bad humidification on fuel cell performance and durability have been published [4,10,124,125]. These studies show that humidification process is a key point in fuel cell durability improvement and a bad humidification control can lead to membrane destruction. It is then relevant to think that a failure in the humidifier and humidification process could lead to fuel cell stack destruction. However, to the better of our knowledge, no aging phenomena and degradation laws have been reported in the literature then, because a lack of data, no degradation model can be developed.

4.2 Power converters

To the better of our knowledge no degradation laws have been reported in the literature. Various studies have exposed different DC/DC converter models but none of them consider performance degradation [33,126–129]. Slah et al. [129] proposed model of a interleaved three-phase dc/dc boost converter dedicated to design and simulation purposes for a fuel cell integrated in an electrical vehicle. The proposed approach is based on an average state-space model which considers the parasitic resistance of MOSFETs and diodes. Zhan et al. [130] presented a model and the associated control strategy of power converters of an uninterruptible power supply with backup integrated in a hybrid fuel cell system. Wu et al. [131] proposed an average model dedicated for simulation of a power conditioning system for a hybrid fuel cell/gas-turbine plant. They made the choice of using an average model instead of detailed model because of the possibility to perform long-time simulation such as hours or days. Besides, as mentioned in the section 1.4.3.4, fault conditions can occur at level of power switches of DC/DC converters. This is why diagnostic algorithm dedicated to fuel cell power converters is an important step without what the complete system can be out of service. Guilbert et al. [23] focused on DC-DC converters and proposed a power switch fault detection tool based on Park's vectors of a 3-leg interleaved boost converter for fuel cell applications. The method has been easily implemented on a FPGA target and experimental results showed that the proposed method is efficient to detect, identify and handle, quickly the faulty leg. Guilbert et al. [36] also proposed a study focusing on the investigation of power switch failures tolerance of a 4-leg floating interleaved DC/DC boost converter. They demonstrated that the loss of one leg in case of open-circuit fault cause additional electrical stresses on power devices and inductors. They also showed that power switch faults lead to an increase of the input current ripple which affect fuel cell lifetime. For this purpose, they proposed a power switch fault detection algorithm and a fault-tolerant control strategy without any additional components. The proposed fault-tolerant control consists in modifying the leg shift of the PWM gate control signals according to the faulty leg. Experimental tests showed that the fuel cell current ripple decreases significantly with the application of the proposed strategy.

4.3 Hydrogen tanks

Some studies focused on failure prediction and damage modelling at level of hydrogen tanks [24]. Zhang et al. [24] proposed an interesting recent comprehensive review focusing on failure analysis and prediction model establishment for type III and IV composite high-pressure hydrogen storage tanks. For instance, Xu et al. [132] proposed a 3D parametric finite element model to predict the damage evolution and failure strength of a composite hydrogen tank cylindric part. An algorithm is proposed to simulate and investigate the progressive damage behavior of composite with increasing internal pressure. Liang et al. [30] investigated impact of thermal effects on failure strength of the composite tank. Micromechanics of failure theory is used to predict the failure initiation at constituent level and material property degradation method is used to account for the post-initial failure behavior of the damaged materials. A developed model is then applied to predict the complex failure behavior of the composite subjected to both thermal and mechanical loadings. Model predictions are verified experimentally and compared to traditional finite element analysis. Satisfying agreements are observed. Melnichuk et al. [31] proposed a general method to estimate cavitation risk for polymer materials, especially for the case of the liner of type IV hydrogen tanks. Molkov et al. [133] proposed a physical model to simulate the thermal behavior of an embedded hydrogen storage tank. The proposed model is validated with fueling experimental data from type III and type IV tanks dedicated to on-board hydrogen storage. Zhu et al. [134] proposed a mathematical model describing the dynamic behavior a Metal Hydride (MH) tank. MH tanks allow a low pressure storage of hydrogen. A database issued from a hybrid fuel cell vehicle recording is used to validate the model coupled to a basic static PEMFC model. Chabane et al. [135] proposed a zero-dimensional physics-based model of metal hydride tank. The proposed methods aims at studying the dynamic heat and mass transfers during desorption process. The purpose is to analyze the thermal-fluidic behavior of this technology of tanks. The proposed method has been verified experimentally thanks to tests carried out on a test bench. Results show good accuracy and a good agreement with experiments. Results also highlight the importance of such a model to optimize energy management strategy when coupling a fuel cell with metal hydride tank. Although this technology of tank presents a very good energy density, its very important mass is still a limitation to a common use. To summarize, either to describe high pressure composite tank or metal hydride tank behavior and failure mechanisms, physics-based models have been preferred. The lack of database and long-term durability test could explain that data-driven modeling approaches have not been really considered so far.

5 Conclusion

Through this review work, it has been seen that many studies have focused on fuel cell stack and batteries degradation and how to model their performance evolution. Degradations mechanisms occurring at the level of all components of a complete hybrid fuel cell system have also been presented. However, it can be seen that degradation of fuel cell ancillaries has been barely investigated and although degradation mechanisms have been identified, the evolution of their performance remains a research track to be explored.

Concerning the modeling of fuel cell stack degradation, many methods have been published in the literature, physical based models, data-driven and some hybrid models. Most of physical based models are quasi static and do not consider dynamic behavior. Data-driven models consider dynamic behavior but require a large amount of data to build the model. Hybrid methods seems to be very promising to combine both advantages while reducing the drawbacks. As degradation laws of fuel cells are not fully mastered yet, black box models such as neural networks are used to learn and forecast the loss of performance.

Thanks to a hybrid model, the behavior at the beginning of life (no degradation) could be reproduced without the need of a large amount of data and, in parallel, a machine learning tool can be performed online to learn and predict performance losses all along the system lifetime.

Concerning the battery, the most suitable modeling method seems to be based on electrical circuit model. They do not require a large amount of data and computational requirement are low. Thanks to such a model the dynamic behavior can be reproduced, and the tool will be able to estimate in real time both health indicators: internal impedance and available capacity according to the aging and operating conditions. Machine learning models provide good accuracy, but they require a large database to perform a qualitative learning phase in order to be able to reproduce the dynamic behavior of the battery. Moreover, these models cannot be used to estimate state of health indicators which can be of particular importance to optimize energy management strategy.

Concerning the balance of plant, physics-based models seem to be the best solution at the moment. Indeed, to the better to our knowledge no long-term durability test has been published. It is relevant to think that failures can occur at all components, but degradation laws and performance evolution have not been reported in the literature. Because of a lack of data and knowledge, degradation models cannot be developed, and physics-based performance models remains the best solution so far. In the future, long-term durability tests could follow the performance evolution of ancillaries' overtime and according to operating conditions in order to investigate the presence of degradation phenomena and their impact on global system performance.

6 Acknowledgements

This project has received funding from the Fuel Cells and Hydrogen 2 Joint Undertaking under Grant Agreement No 875087. This Joint Undertaking receives support from the European Union's Horizon 2020 research and innovation programme, Hydrogen Europe and Hydrogen Europe research

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