A review on model-based diagnosis methodologies for PEMFCs

R. Petrone^{a,b}, Z. Zheng^b, D. Hissel^b, M.C. Péra^b, C. Pianese^a, M. Sorrentino^a, M. Becherif^b, N. Yousfi-Steiner^{b,c}

^a Department of Industrial Engineering, University of Salerno, via ponte don Melillo, 84084 Fisciano (SA), Italy

^b FCLAB Research Federation, FR CNRS 3539, FEMTO-ST/Energy Department, UMR CNRS 6174, University of Franche-Comté, rue Thierry Mieg, 90010 Belfort Cedex, France

^c EIFER, European Institute For Energy Research, Emmy-Noether Strasse 11, 76131 Karlsruhe, Germany

e-mail: <u>rapetrone@unisa.it</u>

Abstract

The proton exchange membrane fuel cell systems (PEMFC)s are interesting devices for energy conversion. Recent researches are aimed at developing new monitoring and diagnosis techniques; a good management of these systems would allow optimizing the performance and reducing their degradation. The objective of a suitable diagnostic tool is to identify and isolate the different faults that may occur in the system being monitored in real time. Therefore, the main features of computational methods are accuracy, reliability and high computational speed. In order to perform the diagnosis, it is necessary to evaluate different approaches. In this work different model-based approaches are investigated as well as their validation and applications. An overview of different methodologies available in the literature is proposed, which is oriented to help in developing suitable diagnostic tool for PEMFC monitoring and fault detection and isolation (FDI).

Keywords: PEMFC; fault detection isolation; model-based; on-line diagnosis

1. Introduction

In recent years, the energy demand has become one of the most critical issues of the society due to the problems related with the greenhouse gas emissions and the depletion of fossil resources. Hydrogen is therefore playing a more and more important role in energy conversion, and fuel cells are considered as a promising solution.

The PEMFC operation is based on the electro-catalytic reactions, the hydrogen oxidation at the anode and the oxygen reduction at the cathode. Nevertheless these processes are influenced by the system operating conditions and depend on several physical phenomena occurring inside the cells. Among

Abbreviations: AE, acoustic emission; ANFIS, adaptive neuro-fuzzy inference systems; ANN, artificial neural networks; AR, analytical redundancy; ARX, linear auto-regression model with exogenous input; BPNN, back-propagation neural networks; C_{dl}, charge double layer capacitor; CI, current interrupt; CPE, constant phase element; CV, cyclic voltammetry; CVA, canonical variate analysis; DC, direct current; EIS, electrochemical impedance spectroscopy; ENN, Elman recurrent neural networks; FDI, fault detection and isolation; FTA, fault-tree analysis; GANN, genetic algorithm neural networks; GK, Gastofan-Kessel; HT, high temperature; LPV, linear parameter varying; LS, least squares; LT, low temperature; μ -CHP, micro-combined heat and power; MLPNN, multi-layer feed-forward networks; MOESP, multivariable output error state-space identification; N4SID, numerical algorithms for subspace state-space system identification; NLAR, non-linear analytical redundancy; NN, neural networks; PEMFC, proton exchange membrane fuel cell; PSO, particle swarm optimization; RBS, radial bases function; R_{ct}, charge transfer resistance; R_m, membrane resistance; RMSE, root mean square error; RNN, recurrent neural networks; SOFC, solid oxide fuel cell; SOH, state of health; SR, satisfaction rate; SVM, support vector machines; VAF, variance accounted for; VC-theory, Vapnik-Chervonenkis theory; Z, impedance; Z_w, Warburg impedance.

others, improper water management [1], catalyst degradation and fuel starvation [2] may introduce a voltage drop and even reduce the lifetime of a PEMFC. Different papers analyse the PEMFC durability [3,4,5]. In order to detect such degradation phenomena and optimize the system performance the monitoring and diagnosis of PEMFC become a central objective.

This paper proposes an overview of different methodologies for PEMFC's diagnosis presented in the literature. The overall concept of fault diagnosis consists in three essential tasks: fault detection, fault isolation and fault analysis [6,7,8]. The task of fault detection is to track down fault occurrence during the operating phases. Once a fault is detected, the fault isolation procedure starts. Finally fault analysis is performed to determine the type, the magnitude and the causes of the fault (fault isolation). Generally, according to whether an analytical model is needed, two basic types of approaches can be considered: model-based and non model-based. The former methodology is based on the development of a model able to simulate the behaviour of the monitored system. In case of a model-based approach, the fault diagnosis is performed mostly via residual evaluation, followed by a residual inference for possible fault occurrence detection [9], therefore such method is also known as residual-based diagnosis. The non model-based approach allows detecting and identifying the fault through human knowledge or qualitative reasoning techniques based on a set of input and output data.

The paper focuses mainly on investigating various model-based approaches available in literature for PEMFC fault detection and isolation (FDI), and is organised as follows. In section 1 the main principle and classification of the model-based method are introduced. White-box and grey-box models are introduced in section 2. It proposes an overview of different grey-box models aimed to develop an on-line FDI for PEMFC systems. Models have been organised in parameters identification based, observed-based, and parity space methods. In section 3 black-box models based on artificial intelligence methods are investigated. Finally, a conclusion is made to evaluate each of the presented methods.

2. Model-based approach

In model-based approach a mathematical model can be developed to design or to control or even to perform both tasks for the system under study. Particularly, system design entails adopting complex physical multidimensional models. On the other hand, synthesis models directly derived from experiments can represent a more viable solution for control and real-time applications. Usually the physical multidimensional models are also called "white-box", in which a series of algebraic and/or differential equations are present. The solution of these equations allows the characterization of the system behaviour, while ensuring a high genericity of the method; however a high computational effort could be required. Models directly derived from experiments are also known as "black-box". Despite the low computational efforts required by black-box models make them particularly attractive for on-line monitoring, control and diagnosis applications, especially for complex system such as PEMFC [10], their strong dependence on available experiments reduce their genericity. Therefore, "grey-box" approaches combining the advantages of both physical and empirical models might often represent an interesting alternative solution when high genericity is required. In the particular field of fuel cells, 0-D or lumped approaches were proven to be highly effective to enhance real-time control [11,12] and even to perform both model-based system sizing and control strategies' definition [13].

Model-based approach is very common in FDI methodologies due to the availability of enough sensors employed for the control are usually enough to perform the diagnosis and no additional devices are required for the FDI algorithm implementation [14]. A model is a representation of the physical system and for this reason a perfectly accurate model cannot exist. Therefore a check of the

modelling uncertainty influence on a model-based algorithm is always required to verify its robustness. Each model is characterized by different parameters which are often unknown. In order to guarantee the model accuracy these parameters must be properly identified. The choice of the identification method depends on the process type. Isermann [15,16] proposes several methods for linear and non-linear systems, such as least-squared (recursive, non-recursive, squared root filtering, etc.), and dedicated approaches. The implementation of the methods and their mathematical aspects are evaluated as well. According to Isermann [15,16], when an accurate enough model is implemented, the fault detection starts with the generation and the evaluation of the residuals. During the process, the model runs in parallel with the physical system. The residuals are then generated in real time as the difference between the model and the physical system outputs. Consequently they are analysed by the residual treatment and an inferential process performs the isolation [6]. The modelbased fault diagnosis scheme is depicted in figure 1. Once the residuals are generated, a comparison with a set of thresholds is performed. When a residual value is over the threshold, a symptom is detected [17]. A specific correlation associates the symptom to the system component, localizing the fault. The threshold evaluation is a crucial step for the symptom detection. Indeed models are never perfectly accurate and residuals are always affected by uncertainties introduced by measurements and calculations. In order to take into account the performance sensitivity of the diagnostic tool with respect to disturbances [14], a trade-off between accuracy and robustness is required [17]. For this purpose, Escobet [7] introduced an adaptive threshold based method. Indeed they stated that the isolation approach based on binary detection causes information loss. In order to improve the fault isolation, the residual sensitivity to a fault has to be evaluated. It is verified [7] that the residual sensitivity analysis provides both quantitative and qualitative information about the fault influence on the residuals and their sense of variation, thus improving the fault detection. Escobet et al. [7] show that several faults could present the same binary fault matrix, but characterized by different sensitivities. A residual sensitive matrix has been proposed to detect unexpected compressor and temperature controller failures, air leak, flooding and water blocking phenomena. The results evidenced that all the considered faults are detectable, while the application of a binary signature matrix did not guarantee the same results.

Figure 1 Model-based fault diagnosis scheme [Ding SX (2008)][6].

Every model has to be identified and validated before being employed. PEMFCs behaviours are usually evaluated directly through the polarization curves. Nevertheless other techniques can be applied such as the cyclic voltammetry (CV), the current interrupt (CI) or the electrochemical impedance spectroscopy (EIS). The validation procedure is the last step in system modelling and involves the comparison between the results of the simulation and the measurements. It is crucial that the tests refer to new data, which have never been employed for the model identification. A suitable level of confidence has to be set to consider the uncertainty due to measurements and calculations errors. Figure 1 also highlights another significant task to be performed when applying model-based FDI techniques, namely the residual processing and consequent decision on whether a fault occurred in the system or not. Among several methodologies that have been proposed to suitably perform such a task, the fault-tree analysis (FTA) [18] emerges as one of the more effective tools to detect faults as a function of residual-based generated symptoms. An example of application of FTA approach to fuel cells is given in [19], where the complete development of an FDI-oriented FTA for solid oxide fuel cell (SOFC) systems is described. An overview of the application of FTA in fuel cell diagnosis is also proposed by Yousfi-Steiner et al. [20]. Some examples of FTA relative to the degradation mechanisms in PEMFC are also available [1,2].

3. From white-box to grey-box models

Analytical models, also called white-box models, exploit in space differential equations to simulate the system behaviour. These models are usually very accurate and based on theoretical relationships. In PEMFC modelling, Nernst-Planck, Butler-Volmer and Fick's laws are usually exploited to reproduce the charge transports (electrical and ionic) and mass transfers phenomena. Complexity of these models depends on their objective. In fact, characterizing the system behaviour requires very detailed models with complex equations to solve. Therefore in some cases, these models could appear very difficult to implement on-line. White-box models are aimed at system design and FDI algorithms' design and testing. However, simplified models can be considered for control and diagnosis purposes, evaluating only the parameter values relative to FDI. The grey-box models are based on physical laws supported by a priori knowledge (i.e. data), replacing some complex mathematical equations with empirical formula or map tables. Therefore this approach allows solving the computational burden problem of white-box models. For this approach it is possible to classify the models available in literature in three main categories: (i) parameter identification based; (ii) observed-based; (iii) parity space methods. The different approaches are described below.

3.1. Parameter identification models

PEMFC monitoring can be performed through the identification of models' parameters during FC system operations. When the parameters are related to the behaviour of either components or physical phenomena a correlation with the nominal value (in no faulty conditions) can be analysed. In this approach the faults are modelled as system parameters. When the variation of these parameters achieves a certain limit, the correlated fault can be detected and isolated. The parameters are directly estimated on-line. A parameter identification scheme is shown in figure 2.

Figure 2 Parameter identification scheme [Ding SX (2008)][6].

A good example of parameter identification method is proposed in Zeller et al. [21]. The authors developed a quasi-static circuit-based model for on-board monitoring and control. The theoretical voltage is obtained by combining a voltage source (i.e. Nernst potential) and the system losses. Activation and diffusion losses are modelled as two different voltage sources opposite to the Nernst one; while a resistance characterizes the Ohmic losses. During the tests, the data are acquired by the current sweep, and the non-linear least square method is adopted for parameters identification. In order to verify the validity of the identified parameters, a statistical approach has also been developed. Furthermore the paper focuses on the parameter variation analysis in the case of PEMFC degradation, this increase the robustness of the diagnosis tool.

An original model aimed at reproducing the system behaviour during flooding is proposed by Hernandez et al. [22]. The main research objectives are the global modelling and the fault diagnosis. The authors developed an electrical equivalent circuit (see fig. 3) for charge, matter and energy conservation laws' simulation. Gas fluid dynamics is taken into account through the analogies between the pneumatic elements and the electrical components. The model allows studying the gases' compositions and their partial pressure. Vapour saturation, membrane and gas diffusion layers are also simulated. Nevertheless the electrical model is not enough to simulate the system behaviour in extreme conditions. The parameters are identified through a recurrent least squared method, linearizing the system around the operating point in real time. The model has been validated showing a good representation of the system dynamics. Moreover, the flexibility of the approach allows implementing this model in any commercial software dealing with electrical network analysis. Hernandez [22] developed a diagnosis algorithm considering three main types of failures: (i) flooding; (ii) drying; (iii) membrane deterioration.

Figure 3 PEMFC equivalent circuit developed by Hernandez et al. [22].

Another diagnosis technique based on the electrochemical impedance spectroscopy (EIS), which is a powerful technique to monitor the low and high temperature proton exchange membrane fuel cells (LT - HT PEMFC) systems. Different studies in the literature demonstrate the potentialities of this nondestructive testing method as a tool for investigating electrochemical processes and developing a robust parameter identification based diagnosis [23-28]. The EIS is a widespread experimental technique able to characterize the behaviour of an electrochemical system, and therefore allows analysing several phenomena inside the cell and evaluating the system losses. The idea behind the EIS is to analyse the response of the electrochemical device after a sinusoidal perturbation imposed on the system terminals. The perturbation input is a signal of small amplitude, superimposed on the nominal value of the operating current (galvanostatic mode) or voltage (potentiostatic mode). Per each operating condition the perturbation frequency changes within a based range of values, usually for PEMFC the interval is [0.1 Hz – 1 kHz]. The galvanostatic mode is usually preferred for fuel cells. The impedance (Z) is calculated as the ratio between the response and the perturbation, then it is possible to analyse the impedance spectrum moving use of Nyquist and Bode. The obtained impedance spectrum is a function of the operating conditions and any variation leads to a change of the spectrum shape: in the Nyquist plot different arcs can appear as function of the phenomena occurring inside the cell. The impedance spectra can be represented by a typical equivalent circuit model, named Randle's model (see fig. 4). This circuit consists of two resistors, a capacitor and a nonlinear element, known as Warburg's impedance. The system's Ohmic losses are modelled by the first resistance (R_m) . In order to describe the effects of the electrodes' polarization, the Faraday's impedance is also considered, which takes into account both the activation and the diffusion losses. It is made of a resistance (R_{ct}) for the charge transfer modelling and a non-linear Warburg's impedance (Z_w) adopted to reproduce the effects of the mass transfer. The Faraday's impedance is connected in parallel with a capacitor characterizing the charge accumulation phenomena in the double layer (C_{dl}).

Figure 4 Randle's equivalent circuit.

Fouquet et al. [23] study the flooding/drying phenomena during PEMFC operation. Several tests were made observing the system behaviour versus time. Their article focuses on the development of a suitable on-line monitoring technique based on impedance spectroscopy. Experimental results were analysed and an equivalent circuit model was developed to reproduce the impedance spectra. The authors propose a modified Randle's circuit (see fig. 5). The double layer capacitor is replaced by a constant phase element (CPE) able to characterize the porous electrodes' effect. The authors propose a robust fault detection and isolation diagnosis for PEMFC hydration monitoring. They state that isolating the hydration faults is possible by observing the position of the circuital resistance values in a 3-dimensional space.

Figure 5 Randle's model with CPE element adopted by Fouquet et al. [23].

Also Asghari et al. [24] study the PEMFC performance via the EIS technique. Different experiments were conducted to study the performance variations by increasing and decreasing the bipolar plate clamping torque and the temperature; flooding effects were also analysed. An equivalent circuit model has been developed (see fig.6) in order to simulate the impedance arcs in Nyquist plot. The authors estimate each process by observing the variation of the parameter values. The parameter trends versus the current density were also shown. The aim of the paper is to study the effects of PEMFC losses on the impedance spectrum in order to develop a diagnosis tool able to detect and isolate the faults by observing the model parameters variation.

Figure 6 Equivalent circuit proposed by Asghari et al. [24].

In their paper Legros et al. [25] simulate the system behaviour in order to detect flooding. The authors propose two different methodologies, the first one based on EIS, while the second one adopting the acoustic emission (AE) technique. The AE analysis is based on elastic waves theory, and is adopted for non-destructive control. The analysis of the system conditions is carried out in real time, sensing the imposed acoustic waves' propagation. The physical-chemical phenomena occurring inside the cell influence the wave's amplitude, energy, frequency and form. Therefore, through the monitoring of these parameters, the PEMFC characterization is performed by using spectral and multi-parametric analyses. AE outputs are processed by automated statistical techniques, which classify different cluster in a multidimensional space. This technique allows investigating mechanical damages and flooding or drying phenomena. After several tests, both the EIS and the AE results confirm the possibility to monitor the flooding process in the cell. This article states the relevancy of these methodologies in order to develop an innovative non-invasive online diagnosis tool.

Another paper on PEMFC monitoring based on EIS technique is proposed by Narjiss et al. [26]. The authors develop an innovative method for PEMFC performance optimization and on-line fault detection. The small sinusoidal signal is superimposed on the system directly through the DC/DC converter and the control system allows the on-line spectroscopy without any disturbance in the electrical load. The idea is that all the phenomena involving an impedance variation can be monitored detecting and isolating the possible faults. The current and the hygrometry variation effects were analysed. A similar approach is also proposed by Bethoux et al. [27].

Some authors [28-30] suggest a circuit model for high temperature (HT) PEMFC monitoring. These systems operating at temperature of about 160 °C are less sensible to CO poisoning. An interesting paper on HT-PEMFC performance characterization in presence of CO₂ and CO through EIS technique is proposed by Andreasen et al. [28]. Moçotéguy et al. [29] analyses the HT-PEM behaviour through the EIS technique within a frequency range of [20 kHz to 0.1 Hz]. The target of this paper is to propose the results of long term tests for u-CHP applications, this study is also interesting for the development of a diagnosis tool based on EIS monitoring. First tests are focused on system ageing considering pure hydrogen and reformate gas at the anode and oxygen and air at the cathode side. The system performance are evaluated at different current densities, it is shown that the best results were obtained for pure gases. Then the impedance spectra are analysed to evaluate the influence of the ageing. An equivalent circuit able to reproduce the physical behaviour of the system has been proposed in figure 7. This circuit is composed of an Ohmic resistance in series with two resistancecapacitor parallel circuit. The first one reproduces the high frequency loop, and the second one models the low frequency loop where a constant phase element is introduced. As a first result, it is observed that the value of the Ohmic resistance does not change with ageing, nevertheless at high frequencies, the first loop seems to disappear as the ageing proceeds. On the contrary, the low frequency loop and the values of the associated resistances varies with ageing. To this purpose Jespersen et al. [30] focuses the research on parameters' identification including current density, stack temperature and fuels' stoichiometry. The authors specify how the model can ensure a correct fitting at each frequency; at the same time a physical meaning is given and a good adaptability to variations of the operating condition is obtained. This paper aims to analyse the parameters' behaviours at different operating conditions in order to develop a robust diagnosis for HT-PEMFC. Nevertheless several tests and a qualified human interpretation of the identified parameters are still required.

Figure 7 Equivalent circuit proposed by Moçotéguy et al. [29].

3.2. Observer-based models

Observer-based model is one of the most common approaches implemented for model-based diagnosis. In this approach the model is integrated with the system and runs in parallel with it. The feed-forward evolution of residuals allows the development of the FDI. An observer-based diagnosis scheme is reported in figure 8. A great limitation about its on-line application for PEMFC systems is the calculation time required for the non-linear model solution.

Figure 8 Observer-based residual generator scheme [Witczak, M. (2003)][31].

An example of observed-based model for PEMFC diagnosis is proposed by de Lira et al. [32,33], they adopted a FDI scheme based on adaptive threshold. The method has been tested on the industrial Ballard NEXA[®] system by simulating different faults [32]. The developed dynamic model takes also into account the behaviours of the auxiliaries. The physical process modelling is based on mass conservation law, electrochemical, thermodynamic and zero-dimension fluid dynamic principles coupled with empirical equations. A linear parameter varying (LPV) observer with the Luenberger structure is applied for the residual calculation. This methodology allows the system equation linearization and solving the analytical problem in a discrete-time state space. For this purpose, a linear time-varying system is adopted. The diagnosis is developed by comparing the real system online behaviour with the dynamic model response. Fault isolation is performed by checking the Euclidean distance between the observed and the theoretical relative residuals. The use of adaptive threshold guarantees the method robustness in PEMFC diagnosis. A set of possible faults was developed to test the algorithm robustness. Sensor outputs are analysed by testing the faults in: i) system supply pressure, ii) oxygen consumption, iii) stack voltage and iv) speed of the compressor motor. Sensor outputs has been successfully valuated detecting all the offsets. The technique has been evaluated successfully for all the considered faults.

3.3. Parity space methods

Based on state space model for the residual region characterization, parity space methods adopt the parity relations instead of an observer for residual generation. As for the observer-based, the parityspace approach allows linearizing the system in a discrete subspace in order to simplify the computation. The advantage of this methodology is its subspace framework, which is presented in form of linear algebraic equations [6]. This approach for on-line diagnosis of PEMFCs is proposed by Buchholz et al. [10], (figure 9). The paper highlights the complexity of the on-line implementation for model-based approaches, due to the requirement of high amount of measurements and computational efforts for non-linear equation solution. In order to deal with these matters, the authors propose to linearize the physical model in the parity space linear domain. Different subspace identification methods were considered, namely: the numerical algorithms for subspace state-space system identification "N4SID", the multivariable output error state-space identification "MOESP" and the canonical variate analysis (CVA). The CVA method, first introduced by Larimore [34], showed the best compromise between the model accuracy and the numerical stability. The authors consider two different approaches for the FDI development, one based on Kalman Filter and the other on the inverse model. The purpose is to demonstrate the applicability of these two methods. In the first approach, the authors reconstruct the Kalman filter state sequence directly based on the system input/output data. In the second approach, they use the CVA to develop the model. For the purpose of the method implementation all the stack measurable inputs and the mean cell voltage are used. Kalman filter and invers model approaches show how the linear CVA state-space models can be implemented

to estimate the non-measurable inputs. The inverse model approach shows the best results when used for diagnosis, allowing the detection of all the evaluated faults.

Figure 9 Inverse model scheme considered by Buchholz et al. [10].

The parity space method is also considered by Yang et al. [35]. Based on the phenomenological dynamic model developed by Pukrushpan [36], the authors linearize the model and generate the relative subspace. The analytical redundancy (AR) approach is adopted for FDI applications by setting the system parity matrix and generating the residuals. This method allows deriving a mathematical representation of the FCs through an algebraic system of equations. Residuals are generated by comparing the measured quantities with their mathematical representation [6]. Two of the twenty-two residuals calculated through the parity space approach are selected and analysed in the paper, leading to the relative fault matrix. The authors consider only two residuals based on the stack current, voltage and on the compressor over-voltage values. Results confirm that the selected residuals are valid for flooding, drying and compressor fault detection. The paper also focuses on demonstrating the method's validity and its possible improvement by introducing adaptive thresholds for the fault detection. In a recent paper [37] the same authors extend the procedure to a non-linear case. They develop a five-order state representation to simplify the model, which in this case has not to be linearized. In this case, the FDI is performed by adopting the non-linear analytical redundancy (NLAR) approach.

To summarize the descriptions just reported on grey-box models it can be clearly stated that the parameter identification approach shows a good accuracy and genericity. Indeed, by adopting an equivalent circuit model, it is possible to characterize the different electrochemical phenomena involved in a fuel cell, while simplifying the algorithm implementation and reducing the computational time. Moreover, the equivalent circuit approach can be achieved through the electrical network analysis. In literature, many authors highlight the use of the EIS technique for parameter identification [23-27,29,30]. The capability of characterizing and analysing the PEMFC impedance spectra through an equivalent circuit allows realizing the on-line monitoring and developing a suitable FDI. Some papers [7,32,33,35,37] underline the possibility to improve the FDI robustness adopting adaptive thresholds. In fact, the isolation approach based on binary fault matrix seems to cause some information losses in FDI. For this purpose the use of relative fault matrix have been proposed. The table 1 reports a summary of the approaches discussed.

Table 1. Grey-box models applications.

4. Black-box models

The black-box models are based on statistical data-driven approach. The relationships between the system inputs and outputs are not based on physical equations as for analytical models, but are deduced through suitable experimental databases. The experimental data are split in two different sets, one dedicated to the training procedure for the identification of the input/output correlations and one used for the model validation. Implementation for black-box models is well suited for complex non-linear systems such as PEMFCs, where the identification of physical parameters of grey-box models may require high numerical efforts [17]. On the other hand, a large amount of experiments is required for model identification. The models available in the literature for PEMFC black-box modelling are introduced in the next sections.

4.1 Neural network

Inspired by biological neural networks, artificial neural network (ANN) has been proved to be a powerful tool for nonlinear system modelling [38]. Given a set of input and output data, the ANN has the ability to learn and build a non-linear mapping of the system, which provides encouraging solution for modelling of complex systems, especially those without well-known variable relationships. The basic unit of an ANN is called artificial neuron. According to the organization of the neurons, there are three fundamental topologies: single-layer feed-forward networks, multi-layer (MLP) feed-forward networks and recurrent networks [39]. In feed-forward networks, neurons are organized in certain parallel layers; all input signals flow in one direction, from inputs to outputs. As for recurrent ANN, the outputs of some neurons are fed back either to the same neurons or to the neurons in the preceding layers [40]; thus, a dynamic effect is introduced into the computational system by a local memory process. Moreover, by retaining the non-linear mapping features of the static networks, the RNN are suitable for black-box nonlinear dynamic modelling [41,42]. Among various ANNs, the most applied one for PEMFC modelling is the MLP type. An example of a Multi-layer NN (MLPNN) with two hidden layers is depicted in figure 10.

Figure 10.Example of a multi-layer feed-forward neural network [38]. I: inputs, H and H': hidden neurons, O: output neurons, W $_{j,I}^{h}$: weights between hidden neuron j and input I, W $_{k,j}^{h'}$: weights between hidden neuron k and output neuron k, W $_{m,k}^{o}$: weights between hidden neuron k and output neuron m.

Numerous ANN models for various PEMFC systems have been developed in recent years, including both static and dynamic ones. Good agreements between models and actual systems are reported in literature. A multilayer perceptron (MLP) type ANN is established in [38] for modelling a 500 W PEM fuel cell stack. The stack voltage, as the single model output, is predicted by applying four inputs including the stack current, the stack temperature, the hydrogen and the oxygen flows. However, this model is a static one, which means that it can only make prediction in static operating conditions. A dynamic model composed of four parallel network modules is proposed in [43], with each module dealing with a different frequency range of the input signals. A high accuracy is obtained with the maximum difference between experimental results and model outputs less than 2.9%. In Sisworahardjo et al. [44], a dynamic MLPNN is applied to model a 100W PEMFC stack. Two variables stack current and stack temperature are arranged in input layer, while stack voltage, output power and hydrogen flow are as output nodes. Close agreement with the results of the experimental data are observed. In Chang' paper [45], a new approach combining NN built on the basis of genetic algorithm (GANN) and an optimizing method, the Taguchi method for characterizing various control factors in NN model, is proposed to estimate the output voltage of PEMFC. The proposed method is proved to have better performance than GANN without Taguchi method and the back-propagation neural network (BPNN) model.

Based on the models, fault diagnosis can be further performed based on the residuals generated between the model outputs and the experimental results. Yousfi-Steiner et al. [40] applied two individual Elman recurrent neural networks (ENN)s to detect the occurrence of flooding and drying in a PEMFC system. As one type of a recurrent neural network, ENN is first introduced by Elman in 1990. It consists of three layers: input, hidden and output layers. Firstly, the most influential variables in water management are chosen from a fault tree analysis as NN inputs according to human expert knowledge, that are the current, the air inlet flow rate, the stack temperature and the dew point temperature. As the stack voltage indicates the degradation and the pressure drop is a relevant parameter to describe the flooding in an electrode, these two variables are determined as NN outputs. In the next step, threshold values are set to define their normal ranges of variation. A diagnosis decision is finally made based on the threshold to discriminate flooding, drying and normal operations.

Since the physical parameters used in this model can be easily estimated even in an embedded fuel cell system, the proposed method can be adapted to an on-board system. NN models mentioned in this section are further summarized in table 2.

Table 2. NN applied for PEMFCs modelling

Compared with the analytical methods, ANN has the advantage of an excellent non-linear approximation ability and fewer assumptions for model construction [45]. Furthermore, it has a low sensitivity to noise and can be built based on incomplete database [46]. However, usually a large amount of dataset under a wide range of operating conditions is needed. When using MLPNN, the determination of the number of hidden layers and the number of neurons of each layer is also a critical issue.

4.2 Fuzzy logic

The main motivation of applying fuzzy logic to perform fault diagnosis is to deal with the system uncertainties, ambiguities and non-linearities [47]. A fuzzy model maps inputs to outputs by combining three components: if-then rules, membership functions and logical operators i.e. AND and OR [48]. Unlike the neural network, it establishes relationships between inputs and outputs by mimicking the human reasoning. Numeric data are converted into linguistic variables by membership functions which define how well a variable belongs to the output i.e. degree between 0 and 1 [48].

In Kishor and Mohanty [49], fuzzy models are developed for the adaptive prediction of the cathode pressure/oxygen partial pressure, the stack voltage and the hydrogen partial pressure in a 50kW PEMFC system. Most relevant and non-redundant input variables for each module are firstly selected through utilizing mutual information based technique. Then, a Gastofan-Kessel (GK) clustering algorithm, which has an adaptive distance norm and is suitable for detecting clusters of different geometric shapes in the data set, is applied for extracting fuzzy rules from data. An efficient prediction is finally provided at different load conditions, evaluated by two performance indices, the variance accounted for (VAF) and the root mean square error (RMSE). However, dataset for training and testing is obtained from an analytical model of a 50kW PEMFC system constructed in MATLAB/SIMULINK, not from a real operational system.

Fennie et al.[50] aims to predict the state of health (SOH) of PEMFC stacks by developing a fuzzy logic model. Data from two 5 W PEMFC stacks are used for training, and data from the other two are for testing. Both EIS measurements and I/V data are applied for building the fuzzy logic model. By applying a subtractive clustering to find initial membership functions and rules, a three-input and one-output model is developed. An encouraging correct rate of 87% is finally obtained on the test data for detecting three types of health states-drying, flooding and healthy. To further improve the accuracy and the robustness, more complete dataset should be collected and a fine tuning of the model using supervised learning algorithm is needed.

In [17], a fuzzy model with two inputs, the stack voltage and the stack current, and one outputsatisfaction rate (SR) is built for detecting two types of faults in a 500W PEMFC system. The first fault type is the accumulation of nitrogen or water in the anode compartment, and the second fault type is the drying of the membrane. An experimental polarization curve under nominal operating conditions is obtained as the expected nominal operating points. The output of the fuzzy model provides directly an SR, which indicates the degree of the fuel cell system V-I points deviating from the reference static points (1 means the system is in static operating mode). In order to discriminate the two types of faults, a fault decision process based on the threshold value of SR and also that of first time derivative of SR is developed. The proposed method has been validated by experiments. At last, the author has also provided a possible extension of this method for diagnosing a greater number of faults, by defining and tuning one different fuzzy surface per fault.

Compared with ANN which needs precise learning in a broad range of the faults, the design of rules and membership functions in a fuzzy logic model is based on operating experience or expert knowledge [51]. Thus it has the advantage of simplicity and easy implementation. At the same time, since it is based on prior knowledge only, it has the problem of the on-line adjustment. That means when new types of faults are needed to be considered, rules and membership functions have to be rebuilt.

4.3 Adaptive neuro-fuzzy inference systems (ANFIS)

Compared with the previous artificial intelligence methods, ANFIS is still not so popular in PEMFC diagnosis domain. However, numerous models based on it can be found and have demonstrated obvious advantages in the literature [51-53], which could be useful for further developments of model-based diagnosis methodologies.

ANFIS model, as an effective combination of neural network and fuzzy logic, has gained more and more acceptance in the field of non-linear system modelling. As already mentioned, neural network has the limitation that a wide range of data set under different operational conditions is needed, while fuzzy logic depends completely on human expert knowledge. In an ANFIS model, the membership functions and rules of the fuzzy system are defined and optimized by ANN, thus not requiring any prior knowledge of the system [52]. The typical structure of an ANFIS model includes five layers: the fuzzification layer, the rule layer, the normalization layer, the defuzzification layer and the summation neuron layer [51].

Tao et al. [51] mainly focus on thermal management, which is critical for the improvement of PEMFC's performance and lifetime. ANFIS is applied to build a temperature model of PEMFC adopting a neural network identification method. Input variables of the model include flow rates of fuel and air, and the output variable is the stack temperature. Simulation results show its feasibility to establish a non-linear model for complicated system such as PEMFC. In order to realize the system thermal management, a neural-fuzzy controller is further developed by regulating the gas flow rates. Simulation results indicate that by adopting the controller, PEMFC can reach the desired temperature rapidly with small fluctuation.

Another ANFIS is presented in [52], it predicts a PEMFC voltage under different operating conditions. The structure of the proposed model consists of five inputs, two membership functions for each input, 32 rules and one output. The current density, the fuel cell temperature, the anode and cathode humidification temperatures and the operational pressures are set as input variables. The prediction capability of the model is verified under all considered operational conditions by comparing with experimental data. At last, a perspective of combining the ANFIS model with a physical model is made in order to extend the capability of the model when adding new influential input variables.

In Ramos-Paja et al. [53], three fuzzy-ANFIS models are constructed to model both the steady-state and dynamic behaviour of PEMFC and its support system. The first model allows the prediction of the polarization curves depending on the fuel flow ratio and the current while the second one relates to the time constant of the first-order delay model during a current transient. The third one is a double-layer charge effect model. A satisfying performance of the proposed model is observed both in simulation and in experimental results under all the considered current transient conditions.

Since ANFIS models combine the benefits of both ANN and fuzzy system, it has been proven to be a powerful tool for PEMFC health state monitoring. However, its present applications are most off-line ones and most of the models are focused on a single cell. Its further development on real-time diagnosis of larger power PEMFC stack is much desired.

4.4 Support vector machines (SVM)

Another interesting method that has emerged in recent years for black-box modelling is Support Vector Machines (SVM). It was originally developed by Vapnik on solid Vapnik-Chervonenkis theory (VC-theory) foundations, but has been extended to handle regression problems more recently [54,55]. It is a novel and powerful tool based on statistical learning theories [56]. The basic idea of SVM is to map nonlinear data into a higher dimensional linear space which is called feature space. Then, in the feature space linear regression is performed [57]. It is different from the most traditional ANN which is based on the empirical risk minimization principle as the SVM is based on the statistical learning and structure risk minimization principle, thus the quality and the complexity of the SVM will not be influenced by the dimensionality of the input space [58,59].

Recent applications of SVM in PEMFC domain mainly focus on fuel cell/ stack modelling instead of pattern classification. Its characteristics such as a high degree of accuracy in prediction and a powerful nonlinear-system modelling capacity can be found in the literature [55,56, 60]. Although it is still not widespread in fault diagnosis yet, there seems to be an increasing necessity in its further application in PEMFC diagnosis field.

In Zhong et al. paper [55], a black box SVM model of a Ballard MK5-E^{\odot} PEMFC is proposed to predict the cell voltage. The current density and temperature are included in the model as input parameters. An illustration of the model is depicted in figure 11. During the development of the model, a key step is selecting optimal SVM parameters. A cross validation method is used to determine their values. In the end, a high degree of precision is acquired in the voltage prediction with a mean squared error of 0.02% and a squared correlation coefficient of 99.7%. It is worth noting that the proposed model can be further expanded by incorporating other operating parameters due to its high generalization capability. However, the proposed method is an off-line one, and real-time implementation will be considered in future work.

Figure 11 Illustration of a SVM PEMFC model, with inputs-current density *I* and temperature *T*, and output-voltage *U*. Support vectors and weights are decided during training. [Zhong, Z.-D. et al. (2006)][55].

Another non-linear off-line model based on least squares SVM (LS-SVM) method is reported in Li et al. [60]. Compared with SVM, LS-SVM can significantly reduce the computation time while maintaining maximum precision. A SVM-ARX (linear auto-regression model with exogenous input) Hammerstein type model is developed in this paper to describe dynamic characteristics of a 3 kW PEMFC stack. LS-SVM is applied to represent a static nonlinear block in the Hammerstein model, with three inputs (oxygen gas stoichiometry, current, cooling liquid flow rate) and two outputs (hydrogen partial pressure and stack temperature). It applies a radial basis function (RBF) kernel. Output of the Hammerstein model is compared with a dynamic physical model of the stack. Good predicting performance can be observed.

Application of LS-SVM for modelling can also be found in Zhong et al. [56]. The LS-SVM is used as a part of a hybrid model to forecast the voltage behaviour based on stack current and temperature, while another pressure-incremental model concerns the cathode and the anode pressure. A particle

swarm optimization (PSO) algorithm is adopted to obtain automatically the best set of hyperparameters for the LS-SVM model. The LS-SVM model shows better agreement with the experimental results by optimizing PSO algorithm. However, the proposed model has limitations in its performance under significant pressure changes; also, it isn't valid in low humidity or under extremely high current density.

Compared with models based on other artificial intelligences, the SVM model has a good generalization capability and this capability is independent on the input-data dimensionality [55]. Therefore, it could be quite interesting to extend the SVM model for fault diagnosis of multivariate complex system like PEMFC system, once the threshold of the nominal operating conditions is set. A summary of SVM models employed in the literature is shown in table 3. Compared with fuzzy logic, it possesses a high precision while no necessity of prior knowledge is presented. Compared with ANN, it has excellent generalization ability and it is more robust [55,56]. All of these merits make it very promising in further research of PEMFC system.

Table 3. SVM applied for PEMFCs modelling

5. Evaluation of model-based approaches for on-line FDI

When developing a model, the first step is to have a deep understanding of the system. The system behaviour has to be analysed in order to reproduce all the involved physical phenomena with mathematical laws. Sometimes, the system complexity could limit the model application. In fact, although the physical phenomena are well known, it could be difficult to formulate simple relationships for physical process modelling [61]. The choice on the type of model (white, grey, or black-box) is therefore influenced by the modelling purpose.

Table 4. Model-based approach comparison for PEMFC applications.

White-box models are usually employed in many chemical and thermodynamics problems. Partial differential equations are introduced for mass and energy transport phenomena involving radiation, convection, and diffusion processes [61]. After the model structure formulation, the data matching allows the identification of the model parameters which are not known a priori. The PEMFC system operation is influenced by electro-chemical, thermal, and fluid-dynamics phenomena. Theoretical relationships such as Nernst-Planck, Butler-Volmer and Fick's laws are usually adopted to reproduce electronic and ionic transport, and mass transfer phenomena. PEMFC physical models are usually very accurate and show a high genericity as long as the knowledge of the geometry and the materials is available to evaluate the parameters. However, very detailed models require complex equations to be solved and are not suited for on-line estimation. Therefore, developed white-box in PEMFC are usually considered for system understanding, off-line monitoring, and training simulators.

In general, for PEMFC on-line FDI applications grey and black-box models are suited. Introducing the grey-box models, both the advantages of physical knowledge, and data-driven are exploited. In this way, complex differential equations can be replaced with empirical formula, or artificial intelligence structures. These models may simulate static and dynamic, linear and non-linear behaviours, allowing a correct accuracy and genericity. The use of semi-physical models reduces the structure complexity, verifying the on-line implementation requirements. The overview of different grey-box models aimed to develop an on-line FDI for PEMFC systems have been organised in parameters identification based, observed-based, and parity space methods. In parameter identification models, PEMFC monitoring is achieved to reproduce the system voltage and/or impedance. To this purpose, different papers propose a circuit-based approach modelling the electro-chemical phenomena through circuit element. Although

in static models, a series of resistances are usually considered to reproduce all the system losses, dynamic circuit components are considered for the dynamic modelling. All the papers analyse the influence in PEMFC performance and degradation of state and control variables such as stack current and temperature or fuel stoichiometry. Relevant results are available for flooding detection [22,23,25,26]. In order to implement in-situ diagnosis, the parameter sensitivity analysis is required. Moreover, the available algorithms have been tested together with a parametric analysis for different operating conditions. Although these studies allowed the method robustness improvement, many efforts are still required to achieve on-board implementation. The parameter identification based on EIS monitoring [23-27,29,30] seems to be the most suitable for on-line FDI applications. As a matter of fact, the impedance monitoring by EIS allows detecting several electro-chemical variations involved in PEMFC. The basic idea is then to associate each physical phenomenon to an equivalent circuit component and analyse its parameter variations. A suitable technique for EIS on-line implementation has been proposed by Narjis et al. [26]. This paper could be considered the base ground for future development of on-board FDI based on EIS. Other approaches are also considered. Both observer-based [32,33] and parity space methods [10,35,37] allows the system model equation reduction. Physical models are linearized and an observer or a parity space linear domain is introduced for residual calculation. Although the system equations are simplified, these methods could generate several residuals. However due to the high dimensions of PEMFC models these methods are validated only for a set of residuals. Yang et al. highlight their efforts in a recent work [37] for FDI improvements, extending their model applications also in non-linear domain. Some papers [7,32,33,35,37] underline that the isolation approach based on binary detection could causes some information losses in FDI. Therefore in order to improve the method robustness an adaptive threshold method has been proposed. All authors stated that residual sensitivity provides both quantitative and qualitative information about the fault influence on the residual and in their sense of variation. This methodology offers a great contribution to FDI improvement, representing a suitable reference for future developments.

Finally, a relevant contribution for PEMFC FDI development is also given by black-box models. Although black-box models are more suitable for complex non-linear system on-line monitoring, they are less generic. In fact, when system operates in new configurations or it is influenced by external factors, not considered in training procedures, the robustness of these approaches is reduced [61] as they don't allow extrapolation, only interpolation. Neural networks, fuzzy logic, adaptive neuro-fuzzy inference systems, and support vector machines methods applications have been reported in this paper. Artificial neural networks are mostly used in non-linear dynamic modelling [38,40,43-45]. Starting from an input/output data set, ANN learning process allows the system non-linear mapping. Residuals are directly generated comparing model outputs and experimental results with a high accuracy (less than 2.9%) [43]. ANN guarantees an excellent non-linear approximation with a low sensitivity to noise. The main drawback is that the training process needs of a large amount of dataset under a wide operating condition range which collection might be costly and time consuming. Some authors [17,49,50] introduce successfully the fuzzy logic techniques for on-line PEMFC monitoring, especially for flooding detection. This choice is due to fuzzy logic capability to deal with the system uncertainties, miming human reasoning. This methodology is very easy to implement, but the issue of the on-line adjustment in case of new faults' occurrence has to be considered. In order to solve this constraint adaptive neuro-fuzzy inference systems are adopted. ANFIS allows the coupling of the ANN and fuzzy-logic benefits. The fuzzy rules are defined through the ANN approach, and not through a priori knowledge. However these models are suitable for system behaviour prediction and off-line diagnosis. Also support vector machines have a good generalization capability. In fact this

method is based on statistical learning and don't need a prior knowledge. However, only off-line SVM applications have been applied [55].

The different model-based approaches reported in this paper underline that many efforts are still required in PEMFC on-line FDI. In literature non-model based approaches are also available. These methodologies can be knowledge-based or signal-based. In the non-model based approaches, FDI is performed through fault classification and no residuals are generated. Available experimental data are therefore processed and normalized. Then the different features, which are relevant for fault detecting are extracted. These features are analysed in a proper low-dimensional space. Several techniques such as NN, fuzzy logic, ANFIS, and SVM are employed as fault classifier. This is the main difference between black-box models and clustering techniques. While in the first approach, artificial intelligence and statistical techniques are adopted to model the system and generate residuals, in the second one they are used to classify the fault in a feature space

6. Conclusion

A classification of different model-based approaches for PEMFC systems diagnosis has been proposed including white-box, grey-box and black-box models. A suitable model-based diagnostic tool requires an appropriate combination of system physical characterization and fast implementation of the algorithm. The white-box models can be very accurate. The computation of algebraic and/or differential equations allows a correct characterization of the system behaviour involving a high genericity of the method. Nevertheless in some cases, they could be very difficult to implement online. These models are suitable for different purposes such as the system design and fault generation for FDI algorithm test. The grey-box models are introduced, showing a good accuracy and less effort in computation for on-line diagnosis applications. In particular, the parameter identification approaches based on impedance spectra evaluations emerges as a suitable solution for diagnosis. The equivalent circuits developed in this methodology allow characterizing the PEMFC electrochemical phenomena while at the same time can be easily implemented on board. However the high nonlinearity of the problem could introduce many correlations between the model parameters. Therefore, the use of adaptive threshold for FDI has been introduced. Finally, black-box models for PEMFC diagnosis application have been presented. Compared with the above two models, they do not require physical equations, thus allowing to develop faster algorithms able to ensure also a good prediction of the system dynamics behaviours. Moreover, black-box models give a high approximation of nonlinear phenomena. However, these approaches show a lack of genericity due to the fact that the model characterization is directly based on system empirical data. ANFIS and SVM methods could provide a suitable solution to this issue, however their contribution in PEMFC diagnosis are still for off-line applications.

This paper is the first part of a preliminary work aimed to give an overview on diagnosis techniques considered in literature. An overview on non-model based approaches is proposed in the second part. The objective of the present work is to create the base ground for the development of a suitable diagnostic tool for PEMFC on-line applications.

Acknowledgement

The research leading to these results has received funding from the European Union's Seventh Framework Programme (FP7/2007-2013) for the Fuel Cells and Hydrogen Joint Technology Initiative

under grant agreement n° 256673 - project D-CODE (DC/DC COnverter-based Diagnostics for PEM systems). The support of University of Salerno (FARB projects) is also acknowledged.

References

- [1] Yousfi-Steiner N, Moçotéguy P, Candusso D, Hissel D, Hernandez A, Aslanides A. A review on PEM voltage degradation associated with water management: Impacts, influent factors and characterization. Journal of Power Sources 2008; 183:260-74.
- [2] Yousfi-Steiner N, Moçotéguy P, Candusso D, Hissel D. A review on polymer electrolyte membrane fuel cell catalyst degradation and starvation issues: Causes, consequences and diagnostic for mitigation. Journal of Power Sources 2009; 194:130-45.
- [3] Knights SD, Colbow KM, St-Pierre J, Wilkinson DP. Aging mechanisms and lifetime of PEFC and DMFC. Journal of Power Sources 2004; 127:127–34.
- [4] Schmittinger W, Vahidi A. A review of the main parameters influencing long-term performance and durability of PEM fuel cells. Journal of Power Sources 2008; 180:1–14.
- [5] Wu J, Yuan XZ, Martin JJ, Wang H, Zhang J, Shen J, Wu S, Merida W. A review of PEM fuel cell durability: Degradation mechanisms and mitigation strategies. Journal of Power Sources 2008; 184:104–19.
- [6] Ding SX. Model-based fault diagnosis techniques: design schemes, algorithms, and tools. Berlin/Heidelberg: Springer-Verlag; 2008.
- [7] Escobet T, Feroldi D, de Lira S, Puig V, Quevedo J, Riera J, Serra M. Model-based fault diagnosis in PEM fuel cell systems. Journal of Power Sources 2009; 192(1):216-23.
- [8] Arsie I, Di Filippi A, Marra D, Pianese C, Sorrentino M. Fault Tree analysis aimed to design and implement on-field fault detection and isolation schemes for SOFC systems. ASME. 8th International Conference on Fuel Cell Science, Engineering and Tecchnology 2010; 1:1-11.
- [9] Isermann R. Supervision, Fault-Detection and Fault-Diagnosis Methods An Introduction. Control Engineering Practice 1997; 5:639-52.
- [10] Buchholz M, Eswein M, Krebs V. Modelling PEM fuel cell stacks for FDI using linear subspace identification. IEEE. Conference on Control Applications 2008; 341-6.
- [11] Arsie I, Di Domenico A, Pianese C, Sorrentino M. A Multilevel Approach to the Energy Management of an Automotive Polymer Electrolyte Membrane Fuel Cell System. Journal of Fuel Cell Science and Technology 2010; 7:011004-14.
- [12] Giustiniani A, Petrone G, Pianese C, Sorrentino M, Spagnuolo G, Vitelli M. PEM Fuel Cells Control by Means of the Perturb and Observe Technique. 32nd Annual Conference of IEEE Industrial Electronics Society 2006; 4349-54.
- [13] Sorrentino M, Pianese C. Control oriented modeling of solid oxide fuel cell auxiliary power unit for transportation applications. Journal of Fuel Cell Science and Technology 2009; 6:041011.1-041011.12.
- [14] Simani S, Fantuzzi C, Patton RJ. Diagnosis in dynamic system using identification techniques. London: Springer – Verlag; 2002.
- [15] Isermann R. Model Based fault detection and diagnosis- Status and Applications. ELSEVIER. Annual Reviews in Control 2005; 29:71-85.
- [16] Isermann R. Fault diagnosis systems: an introduction from fault detection to fault tolerance. Berlin/ Heidelberg/New York: Springer; 2008.
- [17] Hissel D, Péra M-C, Kauffmann J. Diagnosis of automotive fuel cell power generators. Journal of Power Sources 2004; 128:239–46.
- [18] Majdara A, Wakabayashi T. Component-Based Modeling of Systems for Automated Fault Tree Generation. Reliability Engineering and System Safety 2009; 94:1076–86.

- [19] Arsie I, Di Filippi A, Marra D, Pianese C, Sorrentino M. Fault Tree Analysis Aimed to Design and Implement On-Field Fault Detection and Isolation Schemes for SOFC Systems. ASME. 8th International Conference on Fuel Cell Science, Engineering and Technology 2010; 1:389-99.
- [20] Yousfi Steiner N, Hissel D, Moçotéguy P, Candusso D, Marra D, Pianese C, Sorrentino M. Application of Fault Tree Analysis to Fuel Cell diagnosis. Fuel Cells 2012. Fundamentals and Development of Fuel Cells Conference (FDFC 2011). 12(2):302-9.
- [21] Zeller A, Rallières O, Régnier J, Turpin C. Diagnosis of a hydrogen/air fuel cell by a statistical model-based method. IEEE. Vehicle Power and Propulsion Conference (VPPC) 2010; 1-6.
- [22] Hernandez A, Hissel D. Modeling and Fault Diagnosis of a Polymer Electrolyte Fuel Cell Using Electrical Equivalent Analysis. IEEE. Transactions on Energy Conversion 2010; 25(1):148-60.
- [23] Fouquet N, Doulet C, Nouillant C, Dauphin-Tanguy G, Ould-Bouamama B. Model based PEM fuel cell state-of-health monitoring via ac impedance measurements. Journal of Power Sources 2006; 159(2):905-13.
- [24] Asghari S, Mokmeli A, Samavati M. Study of PEM fuel cell performance by electrochemical impedance spectroscopy. International Journal of Hydrogen Energy 2010; 35(17):9283-90.
- [25] Legros B, Thivel PX, Druart F, Bultel Y, Nogueira R. Diagnosis and modelling of protonexchange-membrane fuel cell via electrochemical-impedance-spectroscopy and Acoustic-Emission measurements. IEEE. 8th International Symposium on Advanced Electromechanical Motion Systems & Electric Drives Joint Symposium 2009; 1-6.
- [26] Narjiss A, Depernet D, Candusso D, Gustin F, Hissel D. Online Diagnosis of PEM Fuel Cell. IEEE. 13th Power Electronics and Motion Control Conference (EPE-PEMC) 2008; 734-39.
- [27] Bethoux O, Hilairet M, Azib T. A new on-line state-of-health monitoring technique dedicated to PEM fuel cell. 35th Annual Conference of the IEEE Industrial Electronics Society 2009; 2745-50.
- [28] Andreasen SJ, Vang JR, Kær SK. High temperature PEM fuel cell performance characterization with CO and CO2 using electrochemical impedance spectroscopy. International Journal of Hydrogen Energy 2011; 36:981530.
- [29] Moçotéguy P, Ludwig B, Scholta J, Barrera R, Ginocchio S. Long Term Testing in Continuous Mode of HT-PEMFC Based H3PO4/PBI Celtec-P MEAs for l-CHP Applications. FUEL CELLS. MEA'08 Conference 2008; 9(4):325–48.
- [30] Jespersen JL, Schaltz E, Kær SK. Electrochemical characterization of a polybenzimidazole-based high temperature proton exchange membrane unit cell. Journal of Power Sources 2009; 191:289– 96.
- [31] Witczak M. Identification and Fault Detection of Non-Linear Dynamic Systems. PhD. Thesis. University of Zielona, Poland: Góra Press; 2003.
- [32] de Lira S, Puig V, Quevedo J. Robust LPV Model-Based Sensor Fault Diagnosis and Estimation for a PEM Fuel Cell System. IEEE. Conference on Control and Fault Tolerant Systems 2010; 819–24.
- [33] de Lira S, Puig V, Quevedo J, Husar A. LPV Model-Based Fault Diagnosis Using Relative Fault Sensitivity Signature Approach in a PEM Fuel Cell. IEEE. 18th Mediterranean Conference on Control & Automation 2010; 1284-89.
- [34] Larimore WE. System Identification, Reduced-Order Filtering and Modeling via Canonical Variate Analysis. IEEE. American Control Conference 1983; 445-51.
- [35] Yang Q, Aitouche A, Bouamama BO. Fault detection and isolation of PEM fuel cell system by analytical redundancy. IEEE. 18th Mediterranean Conference on Control & Automation 2010; 1371-76.
- [36] Pukrushpan J. Modeling and Control of Fuel Cell System and Fuel Processor. Ph.D. thesis. University of Michigan, Ann Arbor; 2003.

- [37] Aitouche A, Yang Q, Bouamama BO. Fault Detection and Isolation of PEM Fuel Cell System based on Nonlinear Analytical Redundancy. The European Physical Journal Applied Physics 2012; 54(2):23408-19.
- [38] S, Hissel D, Péra M-C, Kauffmann J. On-board fuel cell power supply modeling on the basis of neural network methodology. Journal of Power Sources 2003; 124:479–86.
- [39] Ogaji SOT, Singh R, Pilidis P, Diacakis M. Modelling fuel cell performance using artificial intelligence. Journal of Power Sources 2006; 154:192-7.
- [40] Yousfi-Steiner N, Hissel D, Moçotéguy P, Candusso D. Diagnosis of polymer electrolyte fuel cells failure modes (flooding & drying out) by neural networks modelling. International Journal of Hydrogen Energy 2011; 36:3067–75.
- [41] Nørgaard M, Ravn O, Poulsen NL, Hansen LK. Neural Networks for Modelling and Control of Dynamic Systems. London: Springer-Verlag; 2000.
- [42] Arsie I, Pianese C, Sorrentino M. Development and Real-Time Implementation of Recurrent Neural Networks for AFR Prediction and Control. SAE International Journal of Passenger Cars -Electronic and Electrical Systems 2009; 1:403-12.
- [43] S, Hissel D, Pera M-C, Kauffmann JM. A New Modeling Approach of Embedded Fuel-Cell Power Generators Based on Artificial Neural Network. IEEE. Transaction on Industrial Electronics 2008; 55:437–47.
- [44] Sisworahardjo NS, Yalcinoz T, El-Sharkh MY, Alam MS. Neural network model of 100 W portable PEM fuel cell and experimental verification. International Journal of Hydrogen Energy 2010; 35:9104–09.
- [45] Chang K. The optimal design for PEMFC modeling based on Taguchi method and genetic algorithm neural networks. International Journal of Hydrogen Energy 2011; 36:13683-94.
- [46] Chávez-Ramírez AU, Muñoz-Guerrero R, Durón-Torres SM, Ferraro M, Brunaccini G, Sergi F, Antonucci V, Arriaga LG. High power fuel cell simulator based on artificial neural network. International Journal of Hydrogen Energy 2010; 35:12125-33.
- [47] Mechefske CK. Objective machinery fault diagnosis using fuzzy logic. Mechanical Systems and Signal Processing 1998; 12(6):855–62.
- [48] Dash S, Rengaswamy R, Venkatasubramanian V. Fuzzy-logic based trend classification for fault diagnosis of chemical processes. Computers & Chemical Engineering 2003; 27:347–62.
- [49] Kishor N, Mohanty SR. Fuzzy modeling of fuel cell based on mutual information between variables. International Journal of Hydrogen Energy 2010; 35:3620–31.
- [50] Fennie C, Reisner D, Barbetta J, Singh P. Fuzzy Logic-Based State-of-Health Determination of PEM Fuel Cells. Procs. EVS-18 (Berlin, Germany) 2001.
- [51] Tao S, Si-jia Y, Guang-yi C, Xin-jian Z. Modelling and control PEMFC using fuzzy neural networks. Journal of Zhejiang University - Science A 2005; 6(10):1084–89.
- [52] Vural Y, Ingham DB, Pourkashanian M. Performance prediction of a proton exchange membrane fuel cell using the ANFIS model. International Journal of Hydrogen Energy 2009; 34:9181–87.
- [53] Ramos-Paja CA, Romero A, Giral R. Fuzzy-based modelling technique for PEMFC electrical power generation systems emulation. IEEE. Power Electronics (IET), Journals & Magazines 2009; 2(3):241–55.
- [54] Drucker H, Burges CJC, Kaufman L, Smola A. Support Vector Regression Machines. Advances in Neural information processing systems 1997; 155–61.
- [55] Zhong Z-D, Zhu X-J, Cao G-Y. Modeling a PEMFC by a support vector machine. Journal of Power Sources 2006; 160:293–98.
- [56] Zhong Z-D, Zhu X-J, Cao G-Y, Shi J-H. A hybrid multi-variable experimental model for a PEMFC. Journal of Power Sources 2007; 164:746–51.

- [57] Lu J, Zahedi A. Modelling and control of PEMFC based on support vector machine. IEEE. 21st Australasian Universities Power Engineering Conference (AUPEC) 2011; 1–6.
- [58] Suykens JAK, Vandewalle J. Least Squares Support Vector Machine Classifiers. Neural Processing Letters 1999; 9(3):293–300.
- [59] Li X, Cao G, Zhu X-J. Modeling and control of PEMFC based on least squares support vector machines. Energy Conversion and Management 2006; 47(7-8):1032–1050.
- [60] Li C-H, Zhu X-J., Cao G-Y, Sui S, Hu M-R. Identification of the Hammerstein model of a PEMFC stack based on least squares support vector machines. Journal of Power Sources 2008; 175:303–16.
- [61] Czop P, Kost G., Stawik D, Wszotek G. Formulation and identification of First-Principle Data-Driven models. Journal of Achievements in Materials and Manufacturing Engineering 2011; 44(2):179-86.

List of tables

- Table 1. Grey-box models applications.
- Table 2. NN applied for PEMFCs modelling
- Table 3. SVM applied for PEMFCs modelling
- Table 4. Model-based approach comparison for PEMFC applications.

List of figures

- Figure 1. Model-based fault diagnosis scheme [Ding SX (2008)][6].
- Figure 2. Parameter identification scheme [Ding SX (2008)][6].
- Figure 3. PEMFC equivalent circuit developed by Hernandez et al. [22].
- Figure 4. Randle's equivalent circuit.
- Figure 5. Randle's model with CPE element adopted by Fouquet et al. [23].
- Figure 6. Equivalent circuit proposed by Asghari et al. [24].
- Figure 7. Equivalent circuit proposed by Moçotéguy et al. [29].
- Figure 8. Observer-based residual generator scheme [Witczak, M. (2003)][31].

Figure 9. Inverse model scheme considered by Buchholz et al. [10].

Figure 10. Example of a multi-layer feed-forward neural network [38]. I: inputs, H and H': hidden neurons, O: output neurons, W _{j,1}^h: weights between hidden neuron j and input I, W _{k,j}^{h'}: weights between hidden neuron j and hidden neuron k, W _{m,k}^o: weights between hidden neuron k and output neuron m.

Figure 11. Illustration of a SVM PEMFC model, with inputs-current density *I* and temperature *T*, and output-voltage *U*. Support vectors and weights are decided during training. [Zhong, Z.-D. et al. (2006)][55].

Methods	Papers	Description	Applications	Advantages	Future
Parameter identification	Circuit-based [21]	Quasi-static non- linear circuit model. Algorithm robustness tested imposing a random set of parameters.	On-board monitoring and control for transport applications. Degradation analysis. Predictive maintenance.	Parameters sensitivity to singularity is considered coupling the Minimum Error method with the Occurrence Number method.	developments New tests for accuracy improvements and good confidence index adopting only the Minimum Error method.
	Circuit-based [22]	Charge, matter and energy conservation laws modelled with circuit elements. Kirchhoff's laws adopted for networks analysis. Linearized model.	Real-time applications. PEMFC global modelling. FDI. Flooding detection.	Use of common commercial software for network analysis. Good representation of system dynamics.	Models improvements to take into account also saturated conditions. Identification methodologies improvements. FDI improvements.
	EIS-based [23-27,29,30]	PEMFC impedance modelled with equivalent circuit models. Faults are detected observing the parameter variations.	Linear and non- linear dynamic models. On-line and in-situ monitoring and diagnosis. FDI. Flooding detection. Ageing and degradation analysis.	Non-invasive. Allows associating singular equivalent circuit components for each physical phenomenon. Easy to implement. Robust FDI.	Parameters analysis and improvements of identification algorithms to reduce the method sensitivity to singularity. New tests for on-board FDI.
Observers	[7,32,33]	Adaptive thresholds are performed adopting residuals sensitivity. Fault isolation is performed through geometrical representation.	On-line monitoring and diagnosis. FDI.	Allows adopting analytical methods in a discrete time domain. Allows the residual dynamic evolution monitoring. Good robustness.	Improvement in the isolation process when similarity between different residuals occur. Testing of new residuals for FDI improvement.
Parity space	[10,35,37]	Subspace identification methods are performed adopting the parity relations for residual generation. Adaptive thresholds are used for FDI.	On-line monitoring and diagnosis. FDI.	Allows the analytical system equation reconstruction. Good robustness.	Improvement in the isolation process when similarity between different residuals occur. Extension to the Non-linear case.

Authors	Input variables	Output variables	NN type	Dynamic/static
, S. et al.	(1)stack current;	(1) stack voltage	MLPNN trained	Static model
(2003) [38]	(2)stack temperature;		with back-	
	(3)hydrogen flow;		propagation	
	(4)oxygen flow			
, S. et al.	(1)stack current;	(1) stack voltage	MLPNN trained	Both dynamic
(2008) [43]	(2)stack temperature;		with back-	and static
	(3)hydrogen flow;		propagation	models
	(4)oxygen flow;			
	(5)air humidity			
Sisworahardjo	(1)stack current; (2)	(1) stack voltage;	MLPNN trained	Dynamic
et al. (2010)	stack temperature	(2) stack power;	with back-	model
[44]		(3) the hydrogen	propagation	
		flow		
Steiner, Y. N. et	(1)stack current, flow	(1) pressure drop	Elman recurrent	Dynamic
al. (2011) [40]	rate, stack and dew	(2) stack voltage	NN	model
	temperature			
Chang, K (2011)	(1) operation	(1) output	MLPNN	Static model
[45]	temperature, (2)oxygen	voltage	constructed on	
	flow rate, (3)hydrogen		basis of genetic	
	flow rate, (4)load		algorithm and	
	current, (5)oxygen and		optimized by the	
	hydrogen pressure		Taguchi method	

Authors	Input variables	Output variables	Applications
Zhong Z-D et al. (2006)	(1) current density	(1)cell voltage	Predict cell voltage of
[55]	(2) cell temperature		a PEMFC
Li C-H. et al. (2008)	(1) oxygen gas	(1)hydrogen partial	Describe dynamic
[60]	stoichiometry	pressure	characteristics of 3
	(2) stack current	(2) stack	kW PEMFC stack
	(3) cooling liquid flow	temperature	
	rate		
Zhong, Z-D. et al.	(1) stack current	(1)cell voltage	Develop a system-
(2007) [56]	(2) stack temperature		level hybrid model of
	(3)cathode pressure		a PEMFC
	(4) anode pressure		
Lu J. and Zahedi A.	(1)stack current	(1) oxygen excess	Air flow control of a
(2011) [57]	(2)compressor voltage	ratio	PEMFC system
Li, X. et al. (2006) [59]	(1) hydrogen flow rate	(1) operating	Stack temperature
	(2) cooling water flow	temperature	control of a 1 kW
	rate		PEMFC stack
	(3) air flow rate		

	White-box	Grey-box	Black-box
Structure complexity	High	Moderate	Low
Accuracy	High	Good	Good
Genericity	High	Good/moderate	Moderate/Low
Processing time	High	Moderate/Low	Low
Physical knowledge	High	Moderate	Low
Data-driven	Low	Moderate	High
Application area	System understanding Off-line diagnosis	On-line FDI	On-line FDI Control
	Training simulators		
Static models	OK	ОК	ОК
Dynamic models	OK	OK	OK
Non-linear response	Good	Good	High
On-line applications	Not indicated	OK	OK







Figure4 Randle's equivalent circuit





Figure6 Equivalent circuit proposed by Asghari et al.



Figure7 Equivalent circuit proposed by Moçotéguy et al.





Figure9 Inverse model scheme considered by Buchholz et al.





