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State of the art of SOFC systems modeling and diagnosis

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Table of Content

1 EXECUTIVE SUMMARY

1.1 Description of the deliverable content and purpose

The objective of the present deliverable report is to review the statue of the art of SOFC models and diagnosis schemes. This review is intended to serve as a basis for the development of models, methods for condition monitoring and diagnosis.

This work task provides an overview of the available literature and their respective results dealing with solid oxide fuel cell (SOFC) modeling and diagnosis. From this study the most suited modeling approaches and methodologies for monitor and diagnosis SOFC systems have been selected. A deep investigation has been performed on the state of art of control-oriented models, in particular on 1-D, 0-D and black box models both of SOFC and SOFC systems. Moreover, a review of the works dealing with the development and application of diagnostic strategies related to SOFC stacks and systems has been performed.

1.2 Brief description of the state of the art and the innovation brought

N/A

1.3 Deviation from objectives

N/A

1.4 If relevant: corrective actions

N/A

1.5 If relevant: Intellectual property rights

N/A

2 Abbreviations

ANN	Artificial Neural Network
CHP	Combined Heat and Power
CV	Control Volume
FSM	Fault Signature Matrix
FTA	Fault Tree Analysis
GT	Gas Turbine
LS-SVM	Least Squares Support Vector Machine
PID	Proportional Integral Derivative
PSO	Particle Swarm Optimization
PEN	Positive Electrolyte Negative
RBF	Radial Basis Function
SOFC	Solid Oxide Fuel Cell

3 Introduction

The objective of the DIAMOND project is to improve the performance and enhancing the endurance of SOFC CHP-systems. By monitoring key parameters of the system and analyzing them using a diagnosis tool the state of health of the system can be assessed and the control strategy adapted.
As a starting point for the developments within the project the current status of the systems modeling and diagnosis needs to be reviewed. In previous EU-funded projects, like Design and GENIUS cell, much knowledge has already been gained.

A modern (i.e. computational / digital) system diagnosis mechanism is typically based on comparing the measured, real online operation of the system to an estimate of the expected operation. The estimate of the expected operation is obtained based on a computational model. Therefore, for instance in the case of SOFCs and SOFC power systems, SOFC models play a key role and much of the diagnostics work and literature focusses on the modeling of SOFC, and are therefore covered, per se, to a high extent in this overview.

4 Control-oriented SOFC modeling - State of Art

In literature the models on SOFCs range from zero-dimensional (0-D) to three-dimensional (3-D) with different features and point to different research objectives. From the viewpoint of model function, 2-D and 3-D modeling is typically concerned with the cell and stack design issues while 0-D and 1-D modeling is aimed at control purposes (at system-level) such as prediction of both the transient and steady-state performance of fuel cell/stack and establishing the optimal operating condition (Braun [1]). Moreover, high dimensional models require information about material properties or electrochemical parameters that are not always available or might be difficult to determine. Even so, high dimensional models are still helpful to learn the operation behaviour of fuel cells of different geometry and very useful for creating training data for black-box modeling.
Physical models are mainly based on the knowledge of physic-chemical characteristics (electrically, chemically and kinematically), thus also called as “white” models. They present a high generalizability level that enables modeling SOFC stacks of different geometric features, but require a high computational effort. In contrast, there is another approach only based on experimental database (no requirement for any physical property), known as the black-box modeling. Black-box models are developed particularly for control-oriented applications, i.e. system monitoring, online control and diagnosis. Nevertheless, the high dependency upon experimental data makes these models less generalizable. Finally, grey-box modeling are partially physical and partially empirical, falling in between white and black-box approaches.
For the research target of the present thesis (i.e. diagnostic and control applications) low dimensional models (0- and 1-D) are more appropriate due to the less computational time in comparison with the high dimensional ones (2- and 3-D). Therefore the literature review on SOFC stack models was focused on 1-D, 0-D (grey-box) and black-box models.

4.1 1-D SOFC modeling

In 1-D model, the fuel cell is usually treated as a set of layers including interconnects, air channel, electrodes, electrolyte and fuel channel (Bove and Ubertini [2]). Both gas composition and flow rate in each channel are assumed to be constant and their mean values are used in the simulation. For planar SOFC, the main dimension corresponds to the gas channel and the direction is determined by the gas flow. It is necessary to note that the fuel cell with cross-flow design cannot be simulated by 1-D models. For tubular SOFC, the kept dimension is usually the tube axis which coincides with the direction of the fuel and oxidant flow (Bove and Ubertini

[3]).

Magistri et al. [4] built a one-dimensional model for tubular SOFC, where the cell coordinate x is the axis of the tube and its origin corresponds to the bottom of the cell. The main hypotheses of the single cell model are: 1) the cell is adiabatic, 2) the cell voltage is uniform and all the chemical reactions within the anodic stream are at equilibrium, 3) the electrochemical reaction of H_2 is taken into consideration; the electrochemical reaction with CO is neglected. The cell model includes: electrochemical performance, equilibrium of reforming and shifting chemical reactions, mass balances at the anode and cathode, energy balances of gases flows, energy balance of the tube and of the solid positive-electrolyte-negative (PEN) structure. In the paper, the 1-D model was described and the results were compared to the 0-D model simulation proposed by Costamagna et al. **Fout! Verwijzingsbron niet gevonden.**. In both models, the input data are: geometrical characteristics, operating conditions, inlet flow conditions and gas and material properties. The models comparison showed that the temperature inside the stack was not uniform and, although the average value was acceptable, the maximum values were too high.

In Gubner et al. [6],[7] a so-called dynamic behaviour model of an SOFC was developed and verified. The model was capable of reproducing the I-V-behaviour and the temperature distribution in the gas flow direction inside a cell operating under either co- or counter-flow mode. It was found to be sufficiently accurate for rapid system simulation (Gubner et al. [8]). The model enabled, e.g., designing the gas flow rates according to the maximum drawn current density and thereby to prevent overheating of cell.

Aguiar et al. [9] developed a 1-D dynamic model for anode supported intermediate temperature planar SOFC with direct internal reforming. This model predicted the SOFC characteristics both in steady-state and in transient operations. It is based on a mass and energy balances and coupled to an electrochemical model. For the mass balance the molar flux in the gas channels in the flow direction was considered. In the fuel channel, three reactions are taken into account: 1) methane steam reforming; 2) water gas-shift; 3) and hydrogen electrochemical oxidation. In the air channel only the reduction reaction of O_2 was considered. In the energy balance were included: the released heat from electrochemical reactions and Ohmic losses; the convective heat transfer between cell components and gas streams; and the in-plane heat conduction through cell components. The thermal flows between the PEN and the interconnect components were supposed to be conductive and radiate. However, in the gas channels, they were assumed to be convective in the gas flow direction and from the gas channels to the solid parts (perpendicular to flow direction).

Jiang et al. [10] set up a 1-D dynamic model for a tubular SOFC with external reforming. The cell was divided into elements along the flow direction. For each element, in the 4 control volumes (CVs) separated along perpendicular axis: the fuel, the solid, the reaction air and the preheated air CVs. Several assumptions were made for the thermal model: 1) for every element, the temperature within each CV was uniform; 2) the radiation and the conduction heat transfer were not taken into account; 3) the convective heat transfer was assumed as the only cause of the temperature gradient in the gas streams. The heat generated for the reactions (shifting, reforming and electrochemical) and the Ohmic losses were computed. The cell voltage at each element was uniform. An equivalent circuit was built to evaluate the influence of the current path length to the Ohmic loss. This model was capable of predicting SOFC characteristics in both the steady and the transient conditions and showed a good reliability. Results from the model showed that high pressure could improve the cell performance whereas higher operating temperature reduced both the Nernst potential and the irreversible losses (Ohmic, activation and concentration).

Sorrentino [11] developed a 1-D steady-state model for co-flow planar SOFC, and extended to counter-flow configuration in Marra [49]. The model was divided into three sub-models: 1) mass balance; 2) energy balance; 3) voltage. The model was based on the control volume approach, according to which the cell was discretized in CVs in the flow direction and divided into three layers: anode channel, cathode channel and cell (solid layer). The cell was assumed to be isopotential and the pressure drop across the fuel and air channels was neglected. The radiative heat transfer and the heat conduction in the solid layer were neglected and the stack was assumed to be adiabatic. The heat convection between solid layer and gas streams and the energy transfer due to the reactants and products were considered dominant in the energy balance. The model showed a good accuracy in the simulation of SOFC states and variables. It was adopted to generate SOFC stack data to be used in a hierarchic modeling approach for control-oriented applications (Sorrentino et al. [12]).

Cheddie et al. [13] upgraded a 0-D real time model to a dynamic 1-D model in order to predict more accurately the temperature and pressure variations along the gas flow direction. The real time capability was maintained by setting up several simplifications: the current density distribution was considered uniform and there was no need to compute the cell current iteratively, thus resulting in a reduction of computational effort. The overpotentials at each node were replaced by the average one across the cell. It was assumed that neither time lag nor dynamic transient occurred in the voltage change after a current variation, so the transient states were not taken into account. The gas concentration was considered dependent only on partial pressure rather than both pressure and temperature. In thermal model, the heat generation was assumed to occur in the PEN only. The heat conduction was negligible in the fluid phase due to the fact that the thermal conductivity is much higher in the solid regions than in the fluid phases. The 1-D model with 21 nodes was proven to require 3.8 ms of computational time for each iteration. The model validation showed that the limiting assumptions did not lead to the significant simulating difference when comparing with a more comprehensive 1-D model without these assumptions.

Kang et al. [14] modified a 1-D dynamic model for a planar SOFC with internal reforming by integrating two simplifications: 1) the PEN, interconnects and gas channels were integrated together along the perpendicular direction, that is, the SOFC is considered to have only one temperature layer; 2) the current density distribution is considered to be uniform within the SOFC, and the cell voltage is determined by the average gas molar fractions and cell temperature. These two simplifications are similar to the assumptions in Cheddie's modeling and by introducing them, the SOFC model was greatly simplified in form.

Bao and Bessler [15] proposed an interesting 1-D+1-D model to simulate the SOFC performance in steady-state condition based on a mix of semi-empirical and analytical models. In the work two different cases are analysed, i.e. isothermal and non-isothermal. About the computational time the model takes 1.3 or 6.7 s to complete a full thermal 1-D+1-D simulation with 20 or 50 along-the-channel control volumes respectively. Even though this model presents a reduced computational time with respect to other 1-D models, it is a steady state model and its performance are not comparable with those of dynamic lumped models, which are however faster.

4.2 0-D (gray-box) SOFC modeling

The main purpose of the 0-D (grey-box) modeling is to develop model-based tools aiming at optimal design, management, control and diagnosis of SOFC units destined to a wide application area (Sorrentino et al. [16]). These models are suitable for massive use when the main characteristics of the system are already available. Therefore the lack of some physical knowledges (i.e. space description) is compensated by introducing other information such as empirical data. Therefore grey-box models mix phenomenological description with simplified assumptions and practical information In lumped models spatial variations are not taken into account (the transformations are considered to define output variables from input ones). In such an approach, the single elements, for instance, compressors, heat exchangers, fuel reformer, partial oxidizers, and contaminant removal apparatus are simulated through independent sub-models (Bove et al. [17]). Furthermore, they allow being easily calibrated and modified for new developed materials.

The grey-box approach is based on a priori knowledge concerning the process and on the mathematical relations which describe the behavior of the system. The starting point is a specific model structure based on physical relations. The construction procedure of a grey-box model based on mathematical relations can be divided into different sub-tasks: basic modeling, conduct experiment of the process, calibration and validation (Sohlberg et al. [18]). In order to optimize design, control and diagnosis of SOFC systems, with particular regard to the management of energy and mass flows during system start-up and load changes, it is important to simulate SOFCs in transient conditions. The development of these models should meet the compromise between satisfactory accuracy and affordable computational burden. The above compromise can be easily achieved by 0-D (i.e. lumped) modeling approaches Bhattacharyya et al. [19].

Costamagna et al. **Fout! Verwijzingsbron niet gevonden.** described a hybrid system where the SOFC was simulated with the 0-D model approach. The balance equations were written as macroscopic balances, in form of finite equations. The equations expressed a balance between inlet and outlet flows of mass and energy in each component of the system; under suitable assumptions, they allowed the evaluation of the average values of the physical-chemical variables of each components and the electrochemical performance of the group itself.

In Campanari's 0-D SOFC model [20] the cell voltage was a function of the current density, the operating temperature and pressure as well as the reactants and product composition.

Bove et al. [21] built a macro model in which the Ohmic polarization depends upon the material properties only. The open circuit voltage and the activation polarization were related to gas concentration while the concentration polarization was ignored. The mean current density was regarded as an input variable.

Ferrari et al. [22] and Magistri et al.[23] made a transient analysis of hybrid system based on SOFC. This system was mainly composed of three parts: the stack; the anodic recirculation system with fuel feeding and the cathodic side (air side) where turbo-machinery; and heat exchangers. These researches allowed a deep investigation of the Fuel Cell Stack integrated with reformer and post-combustor models.

An Interesting lumped approach was followed by Sedghisigarchi and Feliachi [24] for control and stability enhancement of SOFC-based distributed generators (Sedghisigarchi and Feliachi [25]). Nevertheless, in Sedghisigarchi and Feliachi [24] average cell temperature was assumed as state variable, thus not allowing to provide some basic information for balance of plant analysis, such as temperature of exhaust gases.

Sorrentino et al. [12] proposed a hierarchical modeling approach to derive a control-oriented lumped model of planar SOFC. The model proposed is capable of simulating temperature and voltage dynamics as function of the main operating variables (i.e. current density, fuel and air utilizations, inlet and outlet temperatures) accurately. The contribution of Sorrentino et al. [12], differently than Sedghisigarchi et al. [24], does take into account temperature variation across the channels, thus being suitable to perform, at low computational cost, accurate balance of plant analyses, including heat exchangers sizing (Bhattacharyya et al. [19]). Thus, Sorrentino and Pianese [16] proposed to extend the lumped approach presented in (Sorrentino et al. [12]) to the modeling of a fully integrated SOFC-APU (i.e. auxiliary power unit). This latter contribution was also proven to be valid for the development of model-based diagnostics tools for mobile SOFC APUs (Sorrentino et al., 2009 (a)) [16].

The 0-D approach was also applied to transient modeling of tubular SOFC by Hajimolana et al. [26], to develop suited strategies aimed at controlling voltage and cell-tube temperature by properly acting on both temperature and pressure of the inlet air flow.

Sorrentino and Pianese [27] presented a grey-box model of a SOFC unit. The core part of the model is the fuel cell stack, made of planar co-flow SOFCs and surrounded by a number of auxiliary devices, namely air compressor/blower, regulating pressure valves, heat exchangers, pre-reformer and postburner. As a consequence of low thermal dynamics characterizing SOFCs, a lumped-capacity model is proposed to describe the response of fuel cell and heat exchangers to load change.

A 0-D model for the simulation of SOFCs based micro-cogenerative power system, fed by natural gas is described in Arpino et al. [28] who focused their work on the control logic implemented on-board. The model was validate and then used to investigate the generator performance under different operating conditions, as well as to design new experiments. The presented results underline the role of the thermal control strategy on the current performance of SOFC cogenerative power systems.

Barelli et al. [29] developed a dynamic SOFC system model to evaluate the thermo-chemical operating condition. The final purpose was to achieve an optimal dimensioning of the main plant components to guarantee a suitable inertia of the systems and evaluate the system global performance. From the same group, a work on SOFC/GT hybrid system was done [30] for a dynamic analysis during part load operation. Again a lumped dynamic model of the system was used, that model assumes a constant operating temperature.

An interesting dynamic model of a pressurized SOFC/GT system, including the thermal coupling between the different components, consisting of fuel cell stack with combustion zone and balance-of plant components is developed in [31]. In this work the authors demonstrate that the model is able to predict the dynamic operation of a hybrid system.

In [32] the control logic problem of the hybrid system is analyzed and a mathematical model with multi-loop control strategy is implemented making use of a dynamic radial basis function (RBF) neural network and an adaptive PID controller. The results show how the model can effectively regulate both temperature and power of the fuel cell, which can operate in a safety range.

The temperature dynamic analysis of a SOFC/GT system is dealt in [33], where a least squares support vector machine (LS-SVM) identification model based on an improved particle swarm optimization (PSO) algorithm is presented. Moreover, this model is compared with the back propagation neural network approach showing a higher prediction accuracy and a faster numerical convergence.

Nanaeda et al. [34] proposed a dynamic model of a SOFC-CHP system, in order to understand system operating limits and improve flexibility, according to end-user needs and system efficiency.

4.3 Black-box SOFC modeling

Most of existing models (1-D, 0-D) are based on physical conversion laws and governing equations (Wang et al. [35]; Yakabe et al. [36]; Recknagle et al.[37]; Xue et al. [38]). Although being useful for design analysis and optimization of SOFC, they are too complex for control and diagnosis of SOC system. This drawback impelled some researchers to attempt black-box methods (Arriagada et al., [39]; Chakraborty [40]; Entchev et al., [41]; Goldberg [42]; Huo et al., [43],[44]; Jang [45]; Milewski et al. [46]). The black-box are input-output (i.e. mapping) models, derived through statistical data-driven approach. Contrary to physical models, they are not based on explicit physical equations but use large databases with experimental data, which represent the behaviour of the system as function of different operating, control and state variables. Any black-box model is built without exploiting any physical law but use only a set of input-output pairs for training procedure is used, instead. Black-box models range from classical regression based approaches to complex artificial intelligence based ones (e.g. Neural Network). It has been demonstrated that the black-box models based on artificial intelligent approaches are very suitable for non-linear systems (Patan [47]). However, such models require a large amount of experimental data (i.e. training examples), which should well represent the behaviour of the system; therefore, the experimental burden for collecting meaningful data may become excessive. Although the experimental load is the main drawback of artificial intelligence-based modeling techniques, their intrinsic high accuracy represents the most attractive characteristic. These two opposite features lead to the main trade-off to deal with when approaching the modeling problem to be solved.

Arriagada et al. [39] proposed a non-linear fuel cell model by using artificial neural networks (ANNs) for evaluating SOFC performance; their model is a two-layer feed-forward network whose outputs are air flow, current density, temperatures of outlet air and fuel, average solid and reversible voltage. The model was trained via backpropagation algorithm with a reduced amount of input and correct output data pairs generated by a physical cell model. Comparing the outputs of the ANN model with that of the physical model, the average values of the errors are well below 1% and the maximum below 4%. Besides the accuracy, the ANN models are much faster and easier to use and suitable for the generation of performance maps.

Milewski et al. [46] used the same ANN structure of Arriagada et al. [39] to simulate the SOFC behaviour (they exploited experimental data for training and testing process). This SOFC model predicts the output cell voltage making use of 9 input parameters (current density, cathode inlet O_2 and N_2 flow densities, anode H_2 and He flow density, anode thickness, anode porosity, electrolyte thickness and electrolyte temperature). A hyperbolic tangent sigmoid transfer function was used as the neuron activation function in the first layer, whereas a linear transfer function was used in the output layer. The testing results show that the ANN can be successfully used in modeling the single solid oxide fuel cell. However, its practical development suffered from some drawbacks such as the existence of local minima in the cost function to be minimized during parameter identification and over-fitting.

Marra et al. [48] developed a steady-state model based on artificial neural networks for the simulation of the SOFC stack voltage. The model is able to simulate the SOFC degradation due to aging, considering the time as input to the ANN.

Pohjoranta et al. used an autoregressive model identified from system data and Kalman filtering to create an estimator and model predictive controller for the stack temperature [60,61]. Also Halinen et al. demonstrated the use of a stationary multivariable linear regression estimator with a PID controller to regulate the SOFC stack temperature under disturbances [62].

5 SOFC diagnosis - State of Art

In the available literature, only few works deal with the development and application of diagnostic strategies related to SOFC stacks and systems. Several approaches have been followed to develop the most suitable algorithm for monitoring and detection of either degradation mechanisms and faulty states at stack or system level.

The basic concepts of diagnosis have been investigated and presented by several authors. Among them, Isermann [50], Witczak [51] and Simani and Fantuzzi [52] described the main task of diagnosis (i.e. fault detection, fault isolation and fault identification) deepening the different methodologies that can be considered for each task. The latter two works have a primary attention towards model-based approaches.

Several authors focused on stack degradation. Among them, Larrain et al. [53] developed an SOFC model for the investigation of stack degradation due to interconnect degradation and anode reoxidation potential. They states that the temperature plays the main role as influencing factor on the degradation process. Furthermore, it has been observed that decreasing the operating temperature ease the effects of anode reoxidation, in contrast with interconnections degradation.

Also Virkar [54] investigated stack degradation phenomena. In the presented paper, a mathematical model has been developed to simulate SOFC stack degradation induced by an increase in cells resistance. Several causes have been identified as responsible of this phenomenon, such as the formation of local hot spots inducing material properties and microstructures modification, or fuel or oxidant non-uniform distribution, or also seals degradation and electrode delamination due to thermal cycling.

On one hand, Gemmen and Johnson [55] investigated the link between SOFC system efficiency and degradation, with main focus on system auxiliaries. On the other hand, Barelli et al. [56] studied the effects of several degradation mechanisms affecting an SOFC and the available diagnostic techniques useful for their identification. They observed that it arduous to univocally identify a specific mechanism through the only analysis of the output stack voltage at a specific operating condition, since several mechanisms may induce equivalence effects on the stack performance. For this reason, advanced diagnostic techniques are required.

Concerning SOFC system fault detection and isolation, in the work of Arsie et al. [57] a Fault Signature Matrix (FSM) related to a generic methane-fueled SOFC system has been developed following a Fault Tree Analysis (FTA). The authors focused on the main components which can be affected by faults, that are the SOFC stack, the fuel reformer, the air blower, the post-burner and the heat exchanger. Through the FTA, the main system variables affected by the considered faults were identified and a binary correlation was established. All the information obtained during the fault trees development have been gathered into a FSM, which gives a direct correlation among faults and symptoms.

The work carried out by Arsie et al. has been improved by Polverino et al. [58], who simulated through a mathematical model a generic SOFC system in both normal and faulty conditions. The performed simulations allowed to develop improved FSMs taking into account the quantitative influence of the considered faults on the observed variables. This approach allows to improve the robustness of the developed FSMs, and their application on real systems.

Sorce et al. [59] developed a model-based diagnostic algorithm, in which the fault detection is achieved through residuals analysis during simulated faulty states. These states were simulated through their own developed model. The residuals obtained through this approach are collected into fault maps, used as reference for fault detection and isolation. Thus, faults isolation is performed by observing the residuals behaviors during system monitoring and comparing them to the aforementioned fault maps.

The documented demonstrations of system diagnostics in commercial SOFC power systems remain few. Esposito et al. [63,64] and Polverino et al. [65], demonstrated fault diagnostics on the HEXIS Galileo 1000N system but others remain unpublished.

6 Conclusions and future development

In this document a state of art of the modeling approaches and diagnosis methodologies of both SOFC and SOFC systems has been presented. The analysis has been focused on control-oriented modeling and this survey will serve as a basis for the development of models and methods for condition monitoring, diagnosis and advanced control for SOFC systems.

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