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Application of Buckingham π theorem for scaling-up oriented fast modelling of Proton Exchange Membrane Fuel Cell impedance

Luigi Russo, Marco Sorrentino*, Pierpaolo Polverino, Cesare Pianese

Department of Industrial Engineering, University of Salerno, Via Giovanni Paolo II, 132, 84084 Fisciano, SA, Italy

HIGHLIGHTS

- A methodology to reproduce PEMFC impedance is proposed.
- Non-dimensional parameters are defined by exploiting the Buckingham's π theorem.
- Good accuracy in PEMFC impedance prediction is proved.
- The possibility to use this methodology with scaling-up purposes is demonstrated.

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ABSTRACT

This work focuses on the development of a fast PEMFC impedance model, built starting from both physical and geometrical variables. Buckingham's π theorem is proposed to define non-dimensional parameters that allow suitably describing the relationships linking the physical variables involved in the process under-study to the fundamental dimensions. This approach is a useful solution for those problems, whose first principles-based models are not known, difficult to build or computationally unfeasible. The key contributions of the proposed similarity theory-based modelling approach are presented and discussed. The major advantage resides in its straightforward online applicability, thanks to very low computational burden, while preserving good level of accuracy. This makes the model suitable for several purposes, such as design, control, diagnostics, state of health monitoring and prognostics. Experimental data, collected in different operating conditions, have been analysed to demonstrate the capability of the model to reproduce PEMFC impedance at different loads and temperatures. This results in a reduction of the experimental effort for the FCS lab characterization. Moreover, it is highlighted the possibility to use the model with scaling-up purposes to reproduce the full stack impedance from single-cell one, thus supporting FC design and development from lab-to commercial system-scale.

1. Introduction

Among the environmentally friendly energy conversion technologies, one of the most suitable solutions to cope with pollutant emissions and global warming issues are fuel cells, which have attracted the interest of many researchers in the last decades. In particular, the Proton Exchange Membrane Fuel Cell (PEMFC) has proved to be the most suitable for applications between 0.1 and 1000 kW [1]. The main advantages of PEMFCs are the high performance, modularity and potentially pollution-free operation. However, this technology has to meet specific technical requirements to become commercially competitive. US Department of Energy (DOE) and European Institutions have set clear technical targets to be achieved by PEMFC systems with costs and lifetime being the main issues; for stationary and automotive applications 40.000 h and 6000 h of operations are required, respectively ([2,3]).

In order to enhance FCS reliability and lifetime, control and diagnostics must be improved thanks to advanced monitoring algorithms, which must be fast enough to operate on-field to obtain effective advantages. Advanced techniques are required to analyze performance losses caused by degradation mechanisms and the impact of the current operating conditions.

The most effective methods used to characterize the performance of a fuel cell are electrochemical techniques, such as Electrochemical Impedance Spectroscopy (EIS), polarization curves,

* Corresponding author.
E-mail address: msorrentino@unisa.it (M. Sorrentino).

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cyclic voltammetry, AC impedance and current interruption techniques [4]. Among these techniques, EIS is the most suitable to recognize the individual contributions related to voltage drops caused by membrane, charge transfer and mass transfer resistances, both for single cells and overall stack [5]. The EIS technique is used to measure the frequency dependence of the impedance of a fuel cell by applying a small sinusoidal AC voltage (or current) as a perturbation signal to the fuel cell and measuring the current (or voltage) response. This is done at different frequencies for each load to obtain the overall system impedance, often plotted as Nyquist diagram [6]. The applications of EIS in PEMFC studies allows providing electrochemical information about the FCS, which can then help in fuel cell structure optimization and selection of the most appropriate operating conditions. The EIS appears to be the most suitable method for fuel cells on-field monitoring and control, thanks to its peculiar characteristics [7]. Indeed, the main advantage of using EIS resides in the possibility to recognize the contributions of each physical phenomenon (i.e. ohmic losses, charge-transfer and diffusion limitations) to the full impedance thanks to the Equivalent Circuit Modelling (ECM), which consists in modelling the cells using equivalent circuits. Therefore, each contribution can be identified since each circuital parameter represents a physical phenomenon occurring inside the fuel cell [8]. Moreover, the analysis of the ECM parameters may be useful to distinguish between normal and faulty operations [9]. The use of Fractional Order Modelling (FOM), instead, has been proposed as a very suitable approach to model transients and time depending phenomena [10].

Many authors have demonstrated the capability to detect faults using the EIS by recording impedance spectra during fuel cell operation and analysing these data using the ECM approach with diagnostic purposes ([11,12]). The paper of Narjis et al. [13] provides the main references for the development of Fault Detection and Isolation (FDI) based on EIS [7]. Also the use of more physical-based modelling representation of fuel cell impedance has been thoroughly accounted for in the literature. For example, Kulikovsky proposed the analytical model of a the impedance of a PEMFC cathode catalyst layer under poor oxygen transport ([14,15]) and at Open Circuit Voltage in addition to the Gas Diffusion Layer impedance [16]. The work of Cruz-Manzo et al. [17] describes the PEMFC impedance spectra subject to Pt oxidation and H_2 peroxide formation at cathode side, also with the combined use of physical and circuit modelling. On the same line, Cruz-Manzo et al. [17] and Niya et al. [18] developed an impedance model combining physical and ECM approaches, particularly aiming at describing a PEMFC impedance at different operating conditions, varying current densities, temperature and humidity levels. Physics-based PEMFC impedance models were also developed by Setzler and Fuller [19], with the implementation of an oxide growth oxygen reduction reaction kinetic model, and Chevalier et al. [20,21], aiming at PEMFC state-of-health and degradation analysis. The analysis of the cells Output Voltage Signal with Discrete Wavelet Transform (DWT) and Continuous Wavelet Transform (CWT) has been also proposed for fuel cells FDI in the recent literature ([22,23]).

The EIS methodology, however, has its drawbacks. The traditional EIS equipment is costly and cumbersome and requires several minutes to record a single impedance spectrum. Moreover, the diagnosis can be performed once some metrics are derived through EIS signal treatment. This is not a simple task because, in many cases, different operating conditions could lead to the same impedance spectrum; thus, a deep analysis based on analyst experience is necessary. On the other hand, for on-field (i.e. unattended) operations suitable algorithms, such as those based on the ECM [24] or on FOM approaches [10], can be built to process the EIS to derive useful metrics for monitoring and diagnostic applications.

The scientific contribution of the present work is the development of a fast model capable of simulating PEMFC impedance spectra. The model is based on non-dimensional parameters defined by exploiting the Buckingham's π Theorem, which is a key method in the similarity theory field. Particularly, it allows suitably describing the relationships linking the physical variables involved in the process under-study to the fundamental dimensions [25]. The definition of non-dimensional parameters allows reproducing the impedance of different types of system operating at different conditions, thanks to the high generalizability of the model. Therefore, the development of such a model does not require large experimental data; moreover, its simple algorithms need few coding operations, low experimental efforts and are computationally efficient (i.e. fast), allowing easily implementation on low cost hardware as well. This method may be then used for several purposes either for design purposes, by exploiting its scaling-up features, or on-system implementation for on-field use such as on-line applications (e.g. monitoring, diagnostics, control, state of health management and prognostics). The authors have already discussed the significant capabilities associated to the proper exploitation of similarity theory for optimal design and management of innovative energy systems, including fuel cell vehicles ([26–28]). The scaling-up features of the presented method allow using either single-cell or short stack arrangements to characterize the electrochemistry, so as to simulate the behaviour of the full stack. During prototyping phases, this may help in reducing hardware costs (for both test bench and cell materials), as well as consumables. Moreover, it is also beneficial in those problems where the occurrence of degradation and faults should be tested under different operating conditions, i.e., when non-optimal control variables are imposed. It is a matter of fact that to characterize FCS several experimental tests have to be performed under different loads. Moreover, to be able to identify occurring faults, tests in faulty operating conditions have to be performed too. These tests are very degradative and implies FCS fast end-of-life. Thanks to the proposed similarity theory-based modelling approach, the latter issues can be avoided by performing a unique EIS measurement at fixed operating conditions, thus obtaining the behaviour of the FCS under different operating conditions by re-characterizing the non-dimensional parameters.

The model developed following the Buckingham theory is mainly achieved through a data-driven approach, which links measured impedance data and physical models to shape the non-dimensional parameters required for the problem description and generalization.

In the following, the description of the overall model is firstly given, starting from the definition of the non-dimensional parameters towards the description of each involved variable. The design problem is formulated considering both physical and electric variables, which are representative of fuel cell impedance (as detailed in Section 2.1). Therefore, key features of both ECM-based and physics-based methods are accounted for. Nevertheless, it is worth observing that the characterization of these non-dimensional parameters requires the use of impedance data provided by experimental tests. The use of such data allows shaping the fuel cell impedance through the non-dimensional parameters by means of proper physical variables (e.g. temperature, current, water content, fuel utilization, etc.), whose values depend on the specific conditions that have to be simulated. Therefore, the novelty of this approach resides in its structure and development process, which could be considered as black-box if seen from a mathematical point of view (as could be any polynomial regression). However, as said, it gathers properties of both ECM-based and physical-based representations: the impedance layout is given by the measured data required for non-dimensional parameters evaluation (without requiring any ECM model), and the scaling-up process is fulfilled