UNIVERSITY OF SALERNO - UNISA
DEPARTEMENT OF INDUSTRIAL ENGINEERING - DIIN

Dottorato di Ricerca in Ingegneria Meccanica
XII Ciclo N.S. (2011-2013)

UNIVERSITY OF FRANCHE-COMTE - UFC
FEMTO-ST LABORATORY / FCLAB INSTITUT

Ecole Doctorale de l'Université de Franche-Comté :
spécialité Sciences pour l'Ingénieur et Microtechniques

CO-DIRECTION of the PhD. Thesis:

"Electrochemical Impedance Spectroscopy for the on-board diagnosis of PEMFC via on-line identification of Equivalent Circuit Model parameters"

Ing. Raffaele Petrone

Tutors
Ch.mo Prof. Cesare Pianese
Dr. Ing. Marco Sorrentino
M. le Pr. Daniel Hissel
M.me le Pr. Marie-Cécile Péra

Coordinator
Ch.mo Prof. Vincenzo Sergi

ABSTRACT

Proton Exchange Membrane, also named Polymer Electrolyte Membrane fuel cells (PEMFC) are interesting devices for energy conversion. Their development is due to the high efficiency, acceptable power density, quick start-up and good environmental compatibility. On the other hand, reliability cost and durability are the main challenges for PEM fuel cell commercialization. In 2010 the American Department of Energy (DoE) sets a target of 40000 hours for stationary and 5000 hours for automotive applications, respectively. Actually, these standards are considered as the mainly reference in fuel cell research.

Based on electro-catalytic reactions, the PEMFC operation is influenced by system functioning conditions. In case of system operation in abnormal conditions several chemical, mechanical and thermal degradation mechanisms could take place inside the cell. Among other, improper water, thermal and gas managements can introduce a cell voltage drop, thus reducing the system performance. A long-term exposure to these phenomena causes the PEMFC lifetime reduction. Thus, a good system management is one of the primary targets to ensure suitable PEMFC durability. For this purpose, research activities are oriented towards the development of newest advanced monitoring and diagnostic algorithms. The primary goal is monitoring the system operation ensuring a correct system control. Moreover, the diagnostic tool (i.e. both algorithm and sensors) allows the detection of system component malfunctioning; it can isolate one or more faults that may have occurred causing the abnormal behaviour of the system operation.

In common commercial systems the operating variables, such as stack voltage, current and temperature are usually monitored for control purposes. The measured signals are then processed through a control board that provides the right control signals to the ancillary devices for the correct operation. However, usual control strategies are finalised to guarantee the system operation in acceptable conditions only, without taking into account any actions for performance recovery. In this contest, advanced research studies, both at experimental and theoretical levels, may support the development of effective monitoring and diagnostic algorithms. From these algorithms the control actions may also be improved by using the knowledge of the system actual status, which in turn can improve the performance. Therefore, the development of appropriate control strategies, as well as accurate fault detection algorithms, are required to attain a longer lifetime. In this scenario, the

capability to identify in real-time the PEMFCs state-of-health and related degradation mechanisms is one of the main objectives.

This work aims at developing a parameter identification algorithm for on-board fault detection and isolation (FDI) applications based on the electrochemical impedance spectroscopy (EIS) technique. The EIS is a non-invasive experimental technique, usually applied for electrochemical system analysis. This procedure stimulates the main physical phenomena involved in PEMFC. Its use is based on the injection of a sinusoidal signal, which perturbs the system at known frequencies. Then, by analysing the system response it is possible to de-couple different electrochemical processes, isolating the PEMFC losses (i.e Ohmic, kinetic and mass transport). Therefore, the idea is to extrapolate and then exploit the information on PEMFC status, which cannot be directly achieved through the cell voltage drop monitoring. To exploit the information brought by EIS data, an equivalent circuit model (ECM) is considered. This allows accounting for the different electrochemical phenomena occurring inside the cell. In each electrical component of the model one or more parameters can be identified. Through their trends it is possible to monitor the system behaviour, and then, to check the possible fault when the system runs.

This work faces two problems: the on-board implementation of EIS and the on-line model parameter identification. The first topic is related to the measurement reliability, which is influenced by system internal and external factors. On the other hand, the second one is oriented to solve the multi-minima problems involved in the minimization function, which can severely compromise the results of the identification process. Moreover, the work aims at replacing the human experience with a carefully automated strategy for the data analysis implementation. Indeed, in common EIS applications, the expertise of the operator is always required for the data interpretation. The proposed procedure performs the automated selection of both the ECM configuration and proper starting parameters values for the fit.

The interpretation of the identified parameters is made through some regression models, which link the parameter changes to the operating conditions variability. For system monitoring, an ECM simulates the impedance spectrum by using the parameters derived from the regressions evaluated at normal operating conditions. Then, the simulated impedance spectrum and the measured one are compared for evaluating residuals. If residuals are less than a fixed threshold, the system operates in normal conditions, otherwise the on-line parameter identification procedure starts. The fault detection and isolation are performed by comparing the values of the identified parameters with their trends expected for the actual operating conditions. Then, it is possible to detect the occurrence of faults when one or more residuals are above the fixed thresholds. For this purpose, a fault to symptoms matrix for drying out, flooding and air starvation is proposed.

This work has been performed within the D-CODE project (website: https://dcode.eifer.uni-karlsruhe.de) funded under Grant Agreement 256673 of the Fuel Cells and Hydrogen Joint Technology Initiative. Its aim is to develop a diagnostic tool for on-line monitoring and FDI based on EIS. For this purpose the classical EIS single-frequency approach based on small amplitude signal injection has been considered. In the context of the project D-CODE, this thesis deals with two relevant issues: the measurements reliability and the impedance spectra analysis.