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Dynamic Modelling and Experimental Validation of a PEM Fuel Cell System

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The aim of the current work is the experimental validation of a detailed dynamic fuel cell model using a formal model-based approach which is oriented towards optimization and control and it relies on electrochemical equations and mass balances. For this scope a fully automated integrated Hydrogen Fuel Cell Testing Unit was developed and used. Therefore the procedure to condition and reactivate the fuel cell is presented. A parameter estimation procedure was employed based on experimental data generated under various operating conditions. The derived data illustrate the transient response during load changes and model predictions have been found in good agreement with experimental data. The developed model has been implemented in the gPROMS^{TM} modelling environment.

1. Introduction

Fuel Cells (FC) systems are part of a promising environmentally friendly electricity generating technology which can be used for stationary and mobile applications. The existing categories are mainly based on the type of electrolyte and the operating conditions. The choice of the operating region leads to different characteristics for the system regarding its profitability, effectiveness and safety. One type of FC is the Polymer Electrolyte Membrane fuel cell (PEMFC) which is currently considered to be in a relative more developed stage. PEMFC's have high power density, solid electrolyte, long stack life, as well as low degradation due to corrosion. The critical operating parameters are mainly the air and hydrogen feed, the regulation of flow, pressure, heat and water management. This work presents the activation procedure of the single FC which is used to produce data for the validation of the dynamic model. The following section presents the dynamic fuel cell model while the subsequent section describes the activation procedure along with the system setup. The last section presents the model validation procedure based on the comparison of model response to experimental data.

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2. Modelling

A semi-empirical isothermal dynamic model was developed with a theoretical background that takes into account main variables, namely the operating temperature, the partial pressures at the electrodes and the fuel cell current.

2.1 Structural Model Analysis and Assumptions

To model mass transport phenomena along the channels, the gas diffusion layers and through the membrane a five volume approach was adopted. Also a simple semi-empirical for nature voltage calculation equation was used that accounts for temperature and the various voltage losses. Finally in this scheme the thermal dynamics of the fuel cell was integrated. To simplify the modelling and reduce the computation cost the following assumptions are drawn: a) the gases are ideal and uniformly distributed, b) the stack is fed with humidified gases hydrogen and air, c) the temperature is constant and uniform in each experiment, d) for the gas channels only one single pressure point is defined because they have a fixed volume with small lengths. Also the produced water is continuously removed from the cathode flow and the condensed water on the anode is dragged by flow of the unreacted hydrogen. The modelling of the gas diffusion layers and the membrane relies on the equations presented in del Real (2007) and their physical parameters are adjusted to the measurements taken from our system.

2.2 Mass Balances

The model equations consist of the standard material balance of each component as every individual gas follows the ideal gas equation. Therefore mass is described through partial pressure of each gas in the material balances. The mass balance is applied to cathode and the anode channel volumes for the respective species. Also the inlet and outlet flows of the channel and the exchange flow between the gas diffusion layers are described in Ziogou et al (2010). The structure of the model and the mass transport throughout each volume is shown in the Figure 1.

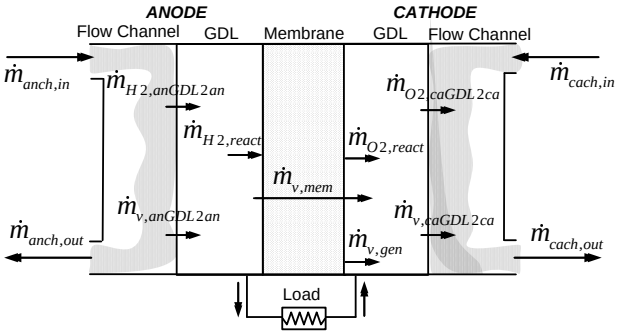


Figure 1: Structure of the Dynamic Model

One important metric of the system is the amount of water vapour in the fuel and air which is calculated from the value of relative humidity, the saturation pressure and the temperature. The equations that give the amount of each species going into the channels which are necessary for the determination of the pressure inside the channels are presented at Pukrushpan et al. (2005). In order to describe the evaporation and condensation dynamics inside the channel, the proposed equations refer to those used by

Golbert and Lewin (2007). The inlet mass flow rate of the nitrogen, oxygen and vapor going into the anode channel are determined in conjunction with the above equations. At the cathode the liquid water condensed is dragged by the air and at the anode the water vapour is dragged by the unreacted hydrogen. The equations that describe the anode channel are developed in an analogous way to the ones describe for the cathode.

2.3 Electrochemical Equations and Voltage Calculation

The description of the fuel cell's behaviour is given through its polarization curve, a plot of voltage versus current density, where the ideal voltage (E_{nerst}) is decreased due to activation (V_{act}), ohmic (V_{ohm}) and concentration losses (V_{conc}):

$$V_{cell} = E_{nernst} - V_{act} - V_{ohm} - V_{conc} \quad (1)$$

The above equation is able to predict the voltage output of various configurations. Depending on the amount of current drawn the output voltage is generated according to equation (1) and the electric power delivered by the system equals the product of the stack voltage V_{cell} and the current drawn I. At very low current density the activation overvoltage appears which takes into account the concentration of oxygen at the catalyst layer (Pathapati et al. 2005). As the current density increases the ohmic losses (V_{ohm}) prevail. They are derived from membrane resistance to transfer protons and from electrical resistance of the electrodes to transfer electrons. Finally the concentration losses (V_{conc}) result from the change in concentration of the reactants at the surface of the electrodes as the fuel is used (Larminie, 2003). The respective equations are:

$$V_{act} = \xi_1 + \xi_2 T + \xi_3 T_{st} \ln(I) + \xi_4 T_{st} \ln(c_{O_2}) \quad (2)$$

$$V_{ohm} = (\xi_5 + \xi_6 T + \xi_7 I) I \quad (3)$$

$$V_{conc} = \xi_8 \exp(\xi_9 I) \quad (4)$$

In the above equations ξ_k*(k=1..9)* represent experimentally defined parametric coefficients whose value can vary from stack to stack.

3. Experimental Validation

The validity of the model was assessed using a single PEM fuel cell that generated required experimental data under various conditions. The effect of temperature, pressure and humidity in the performance of the system was investigated and the validation procedure relies on experiments under both steady state and dynamic conditions. The optimally defined values of the parameters were then used to validate the model under another series of experiments at different operational conditions.

3.1 System Description

In order to study and improve the behaviour and performance of the fuel cell model it is important to measure a variety of variables. Therefore a small scale plant was developed and used. The single PEM fuel cell is integrated with several auxiliary components to form a complete fuel cell system. The Fuel Cell Testing Unit (FCTU) is comprised out of a humidification system, two mass flows for the regulation of the gases and two PID controllers for the anode and cathode pressure regulation. Also the temperature control subsystem includes a fan assisted air cooling system and an electrical heat up system.

This unit was designed based on a modular and flexible architecture and a simplified P&ID of the unit is presented in Fig 2. The voltage and the current density were measured through an on-line supervisory control and data acquisition system (SCADA).

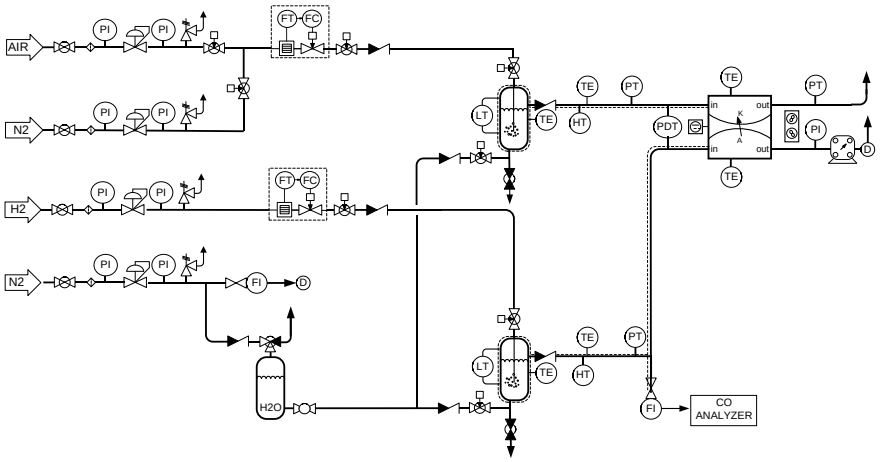


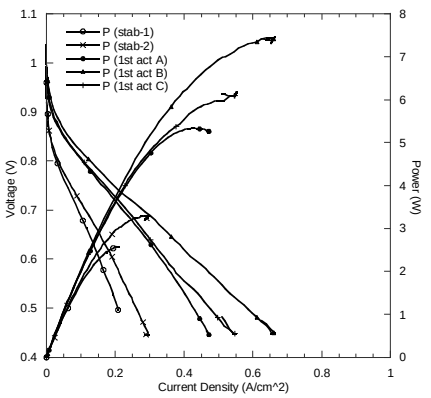
Figure 2: Fuel Cell Testing Unit

The integrated system is equipped also with an electronic load, which simulates the power demands or the power fluctuations that occur in real systems that use fuel cells for power generation. Research in the field of fuel cells demands electronic loads that allow a current to flow even at 0V, as well as load systems with expandable capacity for testing. The electronic load has programmable characteristics depending on the output load required and it is operated in two different modes, the Constant Current (CC) mode and the Constant Voltage (CV) Mode. In CC mode the load current is constant even though the voltage at the terminals of the load changes, while in CV mode the voltage is constant even though the current into the load changes. The activation was performed using the CV mode while the experiments for validation were conducted in CC mode, since the boundaries were determined during full activation procedure. A programming sequence was developed for the load in order to measure the FCs dynamic response.

3.2 Conditioning and Activation

The procedure consists of reading the dynamic response of the cell voltage and cell power after the occurrence of small changes in the load demand, in order to derive the polarization curve throughout the entire operational range. Since the single fuel cell was not conditioned, the system had to be initially stabilized and then the membrane had to be activated through a two stage procedure, the initial activation and the full activation of the membrane. The distinguishing characteristic between these stages is the minimum allowable voltage. Afterwards the performance of the system was tested and the model was validated. The stability of the system was monitored automatically by the control system and when it reached a steady state the load was altered through a series of step changes. A series of small fixed length step changes with a specific range constitute a cycle. The voltage was applied through a ramp procedure and gradually decreased from 1.0 V to 0.50 V and conversely with a step change of 100 mV every 10 s. During the full activation stage the voltage drops to 0 V. The load was constant

during each step. On the following figures the evolution of the current and power density are presented.



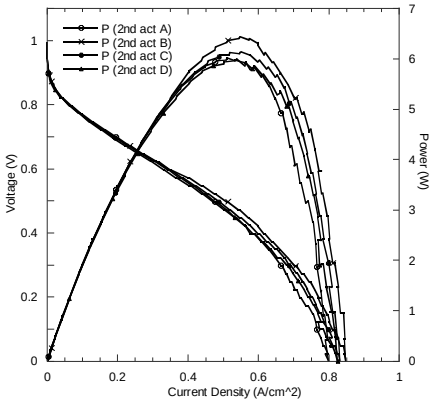


Figure 3a: Initial activation stage (1.0..0.45 V) *Figure 3b: Full activation stage(1.0..0.0 V)*

Through each cycle it was observed that the membrane was further activated. After a series of cycles at the same conditions the membrane presented stable behaviour so that the experimental procedure for the validation of the model could start. Also the system was tested under stressful conditions (elevated pressure, low gas flows) in order to achieve the maximum of its activation.

3.3 Experimental Procedure

The overall experiments are conducted using constant pressure and temperature and fixed intervals for the load demand. More specifically the electronic load is at CC mode and it from 0 A to 20 A with a step change of 2 A every two min. Two sets of experiments were used for the generation of data for the parameter estimation procedure. The varying condition between the set of experiments was the pressure, 0 Barg and 1 Barg. The operational settings of the system are: a) FC temperature stable at 65 °C and the humidification temperature was 75 °C, b) The air flow was 2000 cc/m and the hydrogen flow 500 cc/m. At each condition four experiments were conducted.

3.4 Parameter Estimation

Simulation runs indicated the sensitivity of the system concerning the most critical parameters to be selected for the estimation. Once the model is constructed, estimation is performed to define a set of selected parameters in voltage losses.

Table 1: Parameters for estimation

Parameter	Est. Value	Up Bound	Low Bound	Std. Dev	95% Conf Int
ξ_1	1.3205	1.4	0.954	$1.04 \cdot 10^{-3}$	$2.04 \cdot 10^{-3}$
ξ_7	$7.85 \cdot 10^{-4}$	$4.3 \cdot 10^{-3}$	$1.1 \cdot 10^{-6}$	$4.12 \cdot 10^{-6}$	$8.1 \cdot 10^{-6}$

A nonlinear regression technique with a constant variance model defining a maximum likelihood estimation problem was applied using gPROMS to determine the optimum values for parametric coefficients in activation losses (ξ_1) and in ohmic losses (ξ_7). The bounds for the parameters and the estimated values are presented in Table 1. Model

predictions are in a very good agreement with the experimental data as indicated by the overall standard deviation metric and the confidence interval values shown in Table 1.

3.5 Validation Experiments and Model Behaviour

For the validation of the model a series of experiments were conducted having as varying condition the pressure (0.5 Barg, 1.5 Barg) and it is shown in Fig 4 that the response is similar to the experimental for the whole range of current variation.

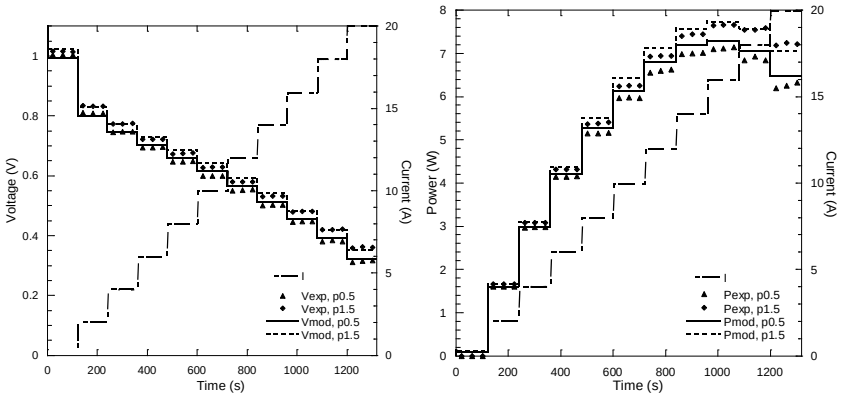


Figure 4: Voltage and power output to load changes for the validated model (0.5Barg, 1.5Barg)

The developed model demonstrated an excellent behaviour both at steady and transient conditions and therefore it can be used both in startup and during variable load changes.

4. Results and Future Work

In this work a dynamic isothermal model was validated using a single cell which was conditioned and used to provide a series of data. The validated model will be the basis for deriving flexible design options and real-time control policies that can be applied to similar experimental units. A parameter estimation procedure was used to determine the optimum values of critical model parameters. Model predictions are in good agreement with experimental data under various operating conditions.

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