Bond Graph for modelling and diagnostics of Proton Exchange Membrane Fuel Cell

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Abstract

Proton Exchange Membrane Fuel Cell is a complex multi-physical system exhibiting complex and coupled phenomenon from several domains of energies (e.g. electro-chemical, electrical, thermal, and hydraulic). However, this promising energy converter is vulnerable to faults (such as drying of the membrane, default of the cooling system and flooding) that can cause permanent damage. Moreover, the performance of the fuel cell is continuously affected by its aging. Therefore, it is essential to implement techniques to detect and isolates those faults. This paper proposes the use of Bond Graph as an integrated tool for the modelling and the generation of fault indicator for this multi-physical system. A diagnostic procedure is also detailed based on the complex structural and dynamical properties in order to isolate a set of single fault.

I. Introduction

Declining fossil energy resources and heavy growth in energy demand has made it imperative to have an efficient energy transition. The use of renewable energy often imposes the long-term storage of electricity. Hydrogen, as an energy vector, can be used for this storage and to be then converted into electricity through a Proton Exchange Membrane Fuel Cell (PEMFC). However, the energy converter suffers from several impairment limiting its industrial deployment [1]. First, a PEMFC has a limited lifespan due to irreversible degradation that are not fully understood [2]. Moreover, it is vulnerable to faults that must be detected early. Among the latter, one can recall the flooding of the channels, drying of the membrane, fault in the cooling system and the loss due to aging. In the past years, the fuel cell community has shown a considerable interest for Fault Detection and Isolation (FDI) in order to ensure the safety when a fault occurs [3-4]. Enhancing the FDI procedure for PEMFC requires a better understanding of the multiple interactions that take place inside the fuel cell stack.

Indeed, a PEMFC is a complex multi-disciplinary energy converter in which several phenomenon are coupled. Its modelling is therefore not a trivial task. Many researches have been conducted to describe its highly non-linear behavior using mechanistic relations [5]. Such models describe accurately the internal physical phenomenon. However, they are not well suited for diagnosis purpose. This is the reason why energy approaches like Bond Graph, are used to model electro-chemical systems (i.e. batteries, super-capacitor) [6]. They aim to describe graphically a device structurally showing the exchanges of power between subsystems with a unified language.

The exploitation of the model structural and causal properties can help for robust fault detection. Indeed, the BG representation allows the systematic generation of Analytical Redundancy Relations (ARR) for the monitorability analysis, for generating the fault signatures with no need for any numerical calculation and then for the diagnostic procedure. The analytical redundancy approach aims at finding the over constrained subsystem exhibiting some redundancies. The first step consists in generating a set of residuals (relations between the known variables of the system) used for the fault detection. The second step is the fault isolation using the generated fault indicators (ARRs) from which is deduced the fault signature matrix.

This paper is organized as follows: section II presents a multiphysical BG model of PEMFC in derivative causality. The third section details the generation of deterministic ARRs from the bond graph model. The monitorability analysis, simulation of considered faults and the associated results are presented in the fourth part. Finally, conclusions and perspectives are discussed in the fifth section.

II. Bond Graph model in derivative causality

The Bond Graph allows to understand the mutual energetic couplings of a multi-physical global system through a simple representation of several subsystems connected to each other with halfarrows showing the exchanged powers (two conjugated power variables: effort and flow). Those power variables have a different signification depending on the considered field of physics. An electrical system involves the pair voltage and current (U,i), the hydraulic field requires the pair pressure-mass flow $(P,\dot{m})$, the chemical and electrochemical reactions are modelled with the pair affinity-speed of reaction (A,J) and the thermal phenomena can be modelled with the pair temperature-enthalpy flow $(T,\dot{H})$ for convection and pair temperature-thermal flow $(T,\dot{Q})$ for conduction [7]. In a real process, the initial conditions can not be known accurately. This is the reason why the derivative causality which is suited for diagnostic, is well suited. All detectors (*De* for the effort and *Df* for the flow) are dualized into sources of signal *SSe* and *SSf* respectively. Following this modelling methodology, the global BG model of a PEMFC is shown in Fig.1 and is detailed in the following sections.

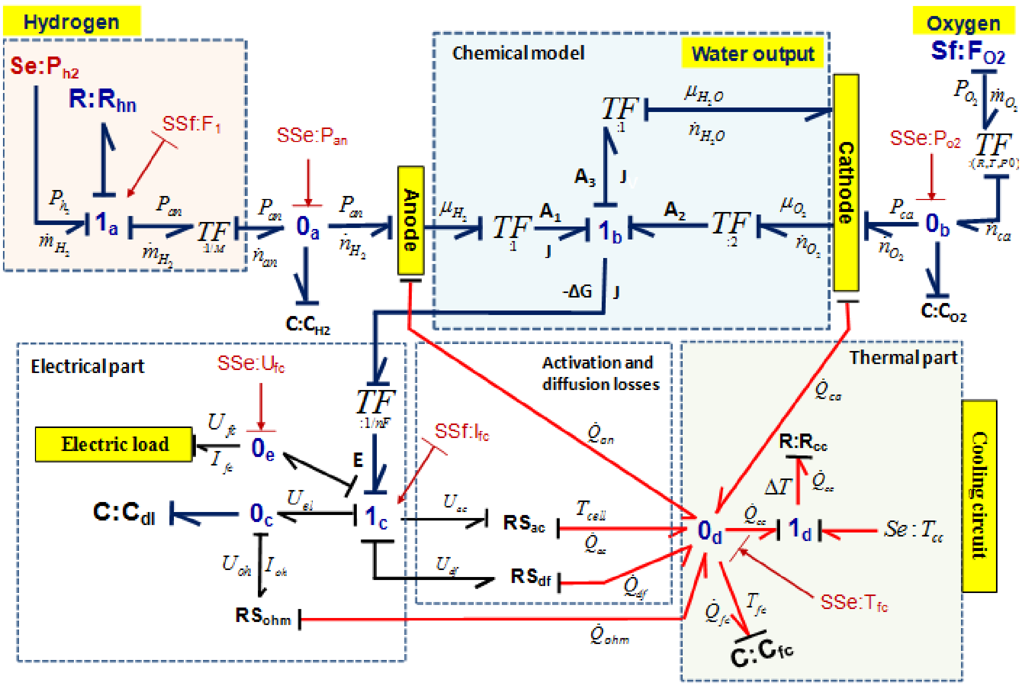

Figure 1. Bond graph model of the PEMFC in preferred derivative causality

II.1. Hydrogen Inlet and Oxygen Inlet

The source of oxygen inlet flow is represented with $Sf:F_{O2}$ while the pressure sensor $SSe:P_{O2}$ measures the pressure on the cathode compartment, the accumulation of gases is usefully represented by a capacitive element $\mathrm{C}:C_{O2}$ and finally a transformer element $\underset{:(R,T,P0)}{TF}$ convert the mass flow into a molar flow. Similarly, $\mathrm{Se}:P_{H_2}$ is the source of hydrogen, the valve is represented using $\mathrm{R}:Rh_n$ and allows to regulates the flow of hydrogen in the anode compartment $SSf:F_{H2}$. The pressure sensor $SSe:P_{an}$ measures the pressure on the anode side and $SSf:F_{H2}$ measures the mass flow rate $\dot{m}_{H_2}$. The accumulation of gases is represented by the capacitive element $\mathrm{C}:C_{H2}$ and a transformer element $\underset{:1/M}{TF}$ allows the transformation of the mass flow into a molar flow (where, M is the modulus representing the molar mass). Finally, the non-linear Bernoulli equation links the flow through the valve $\dot{m}_{H_2}$ and the pressure across the valve P_{Rh} using:

$$P_{Rh} = R_{hn}\left(\dot{m}_{H2}\right)^2 \tag{1}$$

II.2. Chemical Part

At the junction 1_b, the Gibbs free energy $\Delta\mathrm{G}$ models the reaction of reduction-oxidation as:

$$\Delta\mathrm{G}=A_1 + A_2 - A_3 \tag{2}$$

$$\text{with } A_1 = \mu_{H2},\quad A_2 = \frac{1}{2}\mu_{O2},\quad A_3 = \mu_{H2O} \tag{3}$$

$$\mu_{H2} = \mu_0^{H2} + RT_{fc}\ln\left(P_{H2}\right) \tag{4}$$

$$\mu_{O2} = \mu_0^{O2} + RT_{fc}\ln\left(P_{O2}\right) \tag{5}$$

It should be noted that $\mu_{H2O} = \mu_0^{H2O}$ as the water is considered in liquid phase. (6)

μ_i is the chemical potential of specie i and R is the perfect gas constant. In the model, the three transformers $\underset{:\nu_i}{TF}(i=1,2,3)$ are represented through the modulus of the stoichiometric coefficients of the reactant ($\nu_1 = 1$ for hydrogen and $\nu_2 = 2$ for oxygen and $\nu_3 = 1$ for the produced water).

II.3. Electrical and Electro-chemical Part

The chemical subsystem is connected to the electrical and electro-chemical subsystem thanks to the transformer $\underset{:1/nF}{TF}$ that transform the Gibbs free energy into a thermo-dynamical potential E_0 using the following relation

$$E_0 = -\frac{\Delta G}{n_e F} = -\frac{A1 + A2 - A3}{n_e F} \tag{7}$$

where n_e is the number of electrons involved and F is the number of Faraday.

The global ohmic losses (from the membrane, connectors and electrodes) is modelled using a two port thermal dissipative element RS_{ohm} once that heat is generated. Similarly, the activation and diffusion losses are modelled with elements RS_{ac} and RS_{df} respectively, wherein the associated power variables are:

$$U_{ac} = AT_{fc}\, ln\left(\frac{I_{fc}}{I_0}\right) \tag{8}$$

$$U_{df} = BT_{fc} ln\left(1 - \frac{I_{fc}}{I_L}\right) \tag{9}$$

with *A* being the activation constant, *B* being the diffusion constant, I_0 is the exchanged current, I_{fc} is the load current and I_L is the limiting current (maximal current the fuel cell is able to provide).The electrical dynamics (driven by the double layer effect) is modelled using a capacitor element $\mathrm{C}:C_{dl}$. The voltage U_{el} expressed at junction $\mathbf{0}_C$ is the solution of the equation:

$$I_{fc} = \frac{U_{el}}{R_{ohm}} + C_{dl}\frac{dU_{el}}{dt} \tag{10}$$

where R_{ohm} is the global resistance.

II.4. Thermal Part

The chemical reaction (being exothermic) and the active RS elements generate heat that needs to be evacuated. It is chosen to model the thermal part as a first order system, where the thermal resistance and cooling circuit is modelled with a resistive element $\mathrm{R}: R_{cc}$. The thermal dynamic is fixed by the thermal capacitance $\mathrm{C}: C_{fc}$. The cooling water temperature is imposed with an effort source *Se*: T_{cc}. and the temperature of the fuel cell is measured with the sensor *SSe*: T_{fc}. All the power variables are related as:

$$\dot{Q}_{ca} = \Delta S_{H2O}\,\dot{n}_{H2O} - \Delta S_{O2}\,\dot{n}_{O2} \tag{11}$$

$$\dot{Q}_{an} = -\Delta S_{H2}\,\dot{n}_{H2} \tag{12}$$

$$\dot{Q}_{ohm} = R_{ohm} U_{fc} \tag{13}$$

$$\dot{Q}_{fc} = C \frac{dT_{fc}}{dt} \tag{14}$$

$$\dot{Q}_{cc} = R_{cc}\left(T_{fc} - T_{cc}\right) \tag{15}$$

$$\dot{Q}_{df} = \left(R_{df}\right)^2 I_{fc} \tag{16}$$

$$\dot{Q}_{ac} = \left(R_{ac}\right)^2 I_{fc} \tag{17}$$

with $\dot{n}_x$ the molar flow and $\Delta \mathrm{S_x} = \Delta \mathrm{S_{x0}} + \int_{298}^{T} \frac{Cp}{\theta} d\theta$ the flow of entropy of the specie *x*.

III. Generation of determinist ARRs of PEMFC

Analytical Redundancy Relations (ARRs) are the constraint relations derived from the over-constrained system (from the BG model in preferred derivative causality as seen in Fig. 1) and are expressed in terms of measured known variables and system parameters $\boldsymbol{\theta}$ as:

$$ARR: f(SSe, SSf, Se, Sf, MSe, MSf, \boldsymbol{\theta}) \tag{18}$$

The numerical evaluation of the ARR gives a residual $r = Eval[ARR]$. The algorithm to derive the ARRs can be found in [8].

From the junction $\mathbf{0_b}$ (associated with the pressure sensor *SSe* : P_{O2}), the ARR candidate is deduced from the conservative law equation , sum of flows is equal to zero :

$$ARR_1 : \dot{n}_{ca} - \dot{n}_{O2} - \dot{n}_{C_{O2}} = 0 \tag{19}$$

Using the known equations: $\dot{n}_{ca} = \frac{P_{O2}F_{O2}}{RT_{fc}M_{O2}}$ (perfect gas law), $\dot{n}_{O2} = \frac{I_{FC}}{2F}$ (Faraday's law), and $\dot{n}_{C_{O2}} = C_{O2}\frac{dP_{O2}}{dt}$, (19) can be expressed as:

$$ARR_1 = \frac{P_{O2}F_{O2}}{RT_{fc}M_{O2}} - \frac{I_{FC}}{2F} - C_{O2}\frac{dP_{O2}}{dt} = 0 \quad (20)$$

where M_{O2} is the molar mass of oxygen. This ARR is used to monitor a flooding at the cathode compartment. On the anode side, from junction $\mathbf{1_a}$ (associated with flow sensor SSf : F_{H2}), the ARR_2 is deduced:

$$ARR_2 : P_{H_2} - P_{an} - P_{Rh} = 0 \quad (21)$$

Using (1) and known variables, $P_{H2} = Se : P_{H2}$, $P_{an} = SSe : P_{an}$ and $\dot{m}_{H_2} = SSf : F_{H2}$, ARR_2 is expressed:

$$ARR_2 = P_{H2} - P_{an} + R_{hn}\left(F_{H2}\right)^2 \quad (22)$$

This ARR can be used to monitor a fault in the hydrogen valve.

The third ARR is deduced from junction $\mathbf{1_c}$:

$$ARR_3 : E_0 - U_{ac} - U_{df} - U_{el} - U_{fc} = 0 \quad (23)$$

Using the well-known electro-chemical relations [9], the unknown variables can be eliminated using causal paths on the BG model leading to the development of ARR_3 as:

$$ARR_3 = \begin{pmatrix} \mu_0^{H2} + RT_{fc}\ln\left(P_{H2}\right) + \frac{1}{2}\left[\mu_0^{O2} + RT_{fc}\ln\left(P_{O2}\right)\right] - \mu_0^{H2O} \\ -R_{ohm}I_{fc} - AT_{fc}ln\left(\frac{I_{fc}}{I_0}\right) - BTln\left(1 - \frac{I_{fc}}{I_l}\right) \end{pmatrix} - U_{fc} \quad (24)$$

Note that due to fast electrical dynamics, (10) has been approximated as:

$$U_{el} = R_{ohm}.I_{fc} \quad (25)$$

This ARR is sensitive to drying, flooding and to the aging of the fuel cell which is the main focus of the paper. ARR_4 is derived from junction 0_d in the thermal subsystem:

$$ARR_4 : \dot{Q}_{ac} + \dot{Q}_{df} + \dot{Q}_{an} + \dot{Q}_{ca} + \dot{Q}_{ohm} - \dot{Q}_{cc} - \dot{Q}_{fc} = 0 \tag{26}$$

Using the equations (11) to (17), the unknown variables are eliminated:

$$\begin{aligned} ARR_4 = & \left(R_{ac}\right)^2 I_{fc} + \left(R_{df}\right)^2 I_{fc} - R_{cc}\left(T_{fc} - T_{cc}\right) - C\frac{dT_{fc}}{dt} \\ & + R_{ohm} U_{fc} - \Delta S_{H2}\dot{n}_{H2} + \Delta S_{H2O}\frac{I_{FC}}{2F} - \Delta S_{O2}\frac{I_{FC}}{4F} \end{aligned} \tag{27}$$

This ARR can be used to monitor a fault in the cooling system.

IV. Monitorability analysis

For our case, we consider the following phenomena for supervision: drying of the membrane (Dry), water flooding in the cathode channel (Wf), valve failure (Rh), a cooling system failure (Rcc), and aging (Age). In Table I is given fault signature matrix. M_b and I_b are dectability and isolability indexes respectively. They are equal to one if the considered fault is detectable (the signature vector is different from zero) and isolable (signature vector unique) respectively. Regarding the fixed technical specifications, all the faults that may affect the FC system are detectable. However, only those faults that may affect Wf, Rh and Rcc are isolable (their signature vector are unique).

Table I. Fault signature matrix

Ib	0	1	0	1	1
Mb	1	1	1	1	1
ARR/Fi	**Dry**	**Wf**	**Age**	**Rh**	**Rcc**
ARR1	0	1	0	0	0
ARR2	0	0	0	1	0
ARR3	1	1	1	0	0
ARR4	1	1	1	0	1

Aging and drying cannot be isolated as both of them lead to a voltage drop (including ohmic, diffusion and activation losses). To overcome this problem, the decision stage of the algorithm (based on experimental data analysis) developed in [10]. The latter makes use of the first derivative of the fault indicator sensitive to voltage. Once the dynamics of ageing is in the order of few hours and the dynamics of drying phenomenon is in several minutes, the time period during which the failure occurs, remains significantly different. It should be noted that the aging of PEMFC starts from the time it is put into service. As such, once the aging alarm is triggered, the diagnostic procedure will no longer be able to detect the drying of the membrane. This issue has not been considered here and will be addressed in a future work. Nevertheless, the other faults mentioned below are injected and simulated using Matlab Simulink (in order) as:

- Drying: $200 < t < 250$s
- Flooding: $300 < t < 350$s
- Fault in the hydrogen valve: $400 < t < 450$s
- Fault in the cooling system: $500 < t < 550$s

The parameters of the BG model are taken from a previous work [11]. The evaluation of the ARRs gives respectively the four residuals of Fig. 2 (where the thresholds have been chosen arbitrarily).

It can be noticed from Fig. 2 that all the faults are detected and isolated. Nevertheless, the flooding affects the first ARR mainly in the derivative term, causing spikes as fault occurs (not to confuse with a false alarm). The detection of the flooding fault can be improved with a model of the air-compressor where the pressure-flow is monitored or with an observer of the pressure.

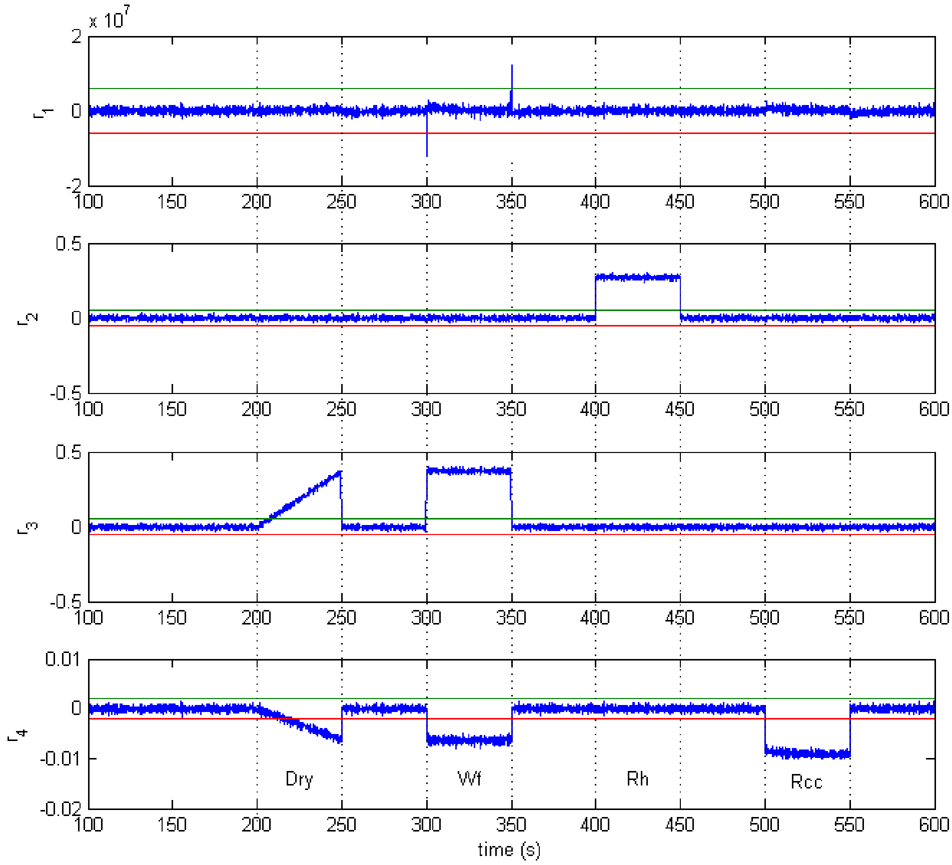

Figure 2. The residuals and their respective thresholds

V. Conclusion

This paper presented a model based diagnostic procedure for PEM fuel cell. First, a multiphysical bond graph model of PEMFC is detailed. The exchanges of power and couplings between subsystems are explicitly shown. Then, four Analytical Redundancy Relations (corresponding to several subsystems) are generated from the BG model. Covering the causal paths, all unknown variables are eliminated and this leads to the monitorability analysis of the system to a set of faults, namely: drying, flooding, aging, fault in the hydrogen valve and fault in the water cooling. Finally, the faulty model is simulated showing the effectiveness of the detection and isolation procedure. Nevertheless, in order to ensure the safe operation of the fuel cell, the flooding must be detected in a better manner before implementing this method in an industrial system. Moreover, the aging that affect the same ARRs than the drying, will always occurs. One can improve the diagnostic procedure by estimating the state of health of the PEMFC using an observer for instance. These issues will be further studied in a future work.

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