Energetic Macroscopic Representation of a Solid Oxide Fuel Cell for Stirling Engine combined cycle in High-efficient Powertrains

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Abstract— **An Energetic Macroscopic Representation (EMR) is developed for a Solid Oxide Fuel Cell (SOFC) in this paper in order to obtain a model well adapted for combined heat and power cycle. Indeed, this study is a first step before coupling this SOFC to a Stirling Engine (SE). Moreover, by using the EMR approach, the possibilities to develop subsequently so-called Maximum Command Structure (MCS) are simplified. Following a first Polymer Electrolyte Membrane Fuel Cell (PEMFC) model established with the EMR formalism, the goal of this study is to adapt this model for SOFC technology and to estimate the heat potential produces by the fuel cell available for the SE hot source. The coupling between the different physical domains encountered in the fuel cell system (fluidic, electrochemical, electrical, and thermal) is represented in a unified manner using the EMR. The final goal of this study is to increase the global electrical efficiency of a coupled SOFC/SE system for automotive applications.**

Keywords*- Solid Oxyde Fuel Cell; Stirling Heat Engine; Combined Heat and Power; Energetic Macroscopic Representation***

Nomenclature—

C_p	Mass heat capacity, $J.kg^{-1}.K^{-1}$
ΔG	Gibbs free enthalpy, $J.mol^{-1}$
ΔH	Reaction enthalpy, $J.mol^{-1}$
ΔS	Reaction entropy, $J.mol^{-1}.K^{-1}$
ΔSq	Entropy flux, $W.K^{-1}$
E	Electrical potential, V
F	Faraday constant, $\mathrm{C.mol^{-1}}$
I	Electrical current, A
M	Molar mass, $kg.mol^{-1}$
n^{el}	Electrons number
$\dot{n}$	Molar flow, $mol.s^{-1}$
p	Partial pressure, Pa
q	Volume flow, $m^3.s^{-1}$
$\dot{Q}$	Heat flow, W
R	Universal gas constant, $\mathrm{J.mol^{-1}.K^{-1}}$
R_i	Electric resistance, Ω
R_d	Fluidic resistance, $Pa.m^{-3}$
S	Tube Cross section, m^2
T	Temperature, K

Greek symbols

α	Charge transfer ratio
ρ	Volume mass, $kg.m^{-3}$

Indices and exponents

0	Reaction standard conditions
C	Consumed
E	Entry
FC	Fuel Cell
i	Gas used
L	Limit
N	Nernst
S	Outflow
x	anode (a) or cathode *(c)*

I. INTRODUCTION

The exponential increase in world energy demand, due especially to actual human way of life, results in geopolitical trends on energetic fossil resources and in an explosion of greenhouse gases (GHG) emissions. Thus, the energy systems conversion efficiency needs to be optimized, especially considering transportation applications (contributing more than 25% to the global GHG emissions).

To increase this efficiency, new technologies have to be applied. For instance, the hybridization of sources (cogeneration) is a way to improve the global efficiency of a given system [1] [2]. To reach this aim, this paper considers the association of Solid Oxide Fuel Cell (SOFC) with a Stirling Engine (SE). The SOFC stack is producing, from a given fuel and air, electricity and, as by-product, high temperature heat (700−800°C). The SE is here used to enhance this resulting heat into electricity, thus contributing to a global improvement

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of electrical efficiency and to a decrease of the size of the requested heat exchangers.

The resulting hybrid power device features nevertheless highly multiphysics behaviour. An efficient modelling formalism intended also to propose control laws must therefore be considered. Energetic Macroscopic Representation (EMR) provides a unified approach for the modelling of multiphysics systems and for the analysis of dynamic systems and is ideally suited for our purpose [3].

Following a first Polymer Electrolyte Membrane Fuel Cell (PEMFC) model established with the EMR formalism [4] [5] [6], the goal of this study is to extend this model for SOFC technology and to estimate the heat produced by the fuel cell stack and available for cogeneration applications.

The first part of this paper will describe the behaviour of a SOFC stack and a SE, as well as the interest of their hybridization. The following part of the paper will then be focused on the EMR of the SOFC stack and will provide the different thermodynamical equations. The experimental validation of the proposed model is given in the last part of the paper.

II. SYSTEM DESCRIPTION

A. *Solid Oxide Fuel Cell*

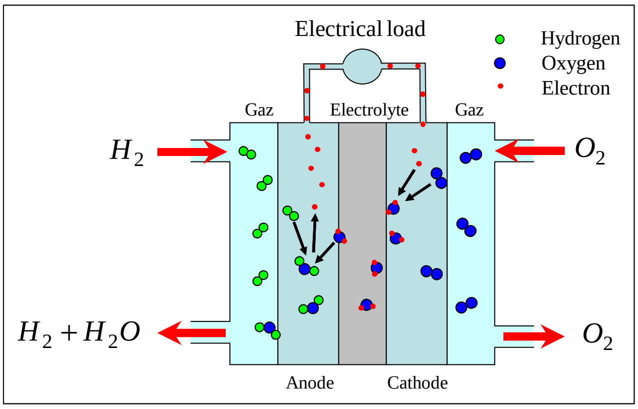

Figure 1: *Electrochemical process of a Solid Oxide Fuel Cell*

As other kind of Fuel Cell (FC) technologies, Solid Oxide Fuel cells are electrochemical devices where the direct conversion of chemical energy (coming from a fuel) into electricity and heat takes place. This kind of FC technology is especially suitable for transportation applications, due to the solid nature of the electrolyte (no leakage risk linked to vibrations). Moreover, their electrical efficiency (up to 60%, considering only the fuel cell stack) is the highest encountered considering the various types of FC technologies [7]. The fuel, in our case hydrogen, is not subject to combustion but to an electro-oxidized reaction EQ. 1 at the anode by oxygen ions moving across the electrolyte FIG. 1.

$$H_2\,(g) + O^{2-} \longrightarrow H_2O\,(g) + 2e- \qquad (1)$$

As shown in FIG. 1, the liberated electrons pass through the electrical external circuit to come back to the cathode where they reduce the oxygen molecule into oxide ions EQ. 2.

$$\tfrac{1}{2}\,O_2\,(g) + 2e^- \longrightarrow O^{2-} \qquad (2)$$

The oxidation of hydrogen follows the reaction EQ. 3. This reaction is exothermic, and heat is a by-product as the production of electricity. Finally the overall reaction of the fuel cell can be written as EQ. 4

$$H_2 + \tfrac{1}{2}\,O_2 \longrightarrow H_2O \qquad (3)$$

$$H_2 + \tfrac{1}{2}\,O_2 \longrightarrow H_2O + electricity + heat \qquad (4)$$

B. *Stirling Engine*

A Stirling Engine (SE) is a thermal engine that can be considered as an externally fired heat engine. It can be powered using a wide variety of fuels and heat sources such as, combustible materials, solar radiation, geothermal hot water, radioisotope energy.

The work fluid hold inside the engine is kept under two heat sources, one hot source and one cold sink. The thermal energy provided by an external hot source (here the by-product of the SOFC stack) is converted into mechanical energy. The theoretical conversion, with a complete regeneration, efficiency EQ. 5 of SE is the Carnot efficiency [9] [10]:

$$\eta = 1 - Tc/Th \qquad (5)$$

where T_c and T_h are respectively cold and hot temperatures of the different engine sources. In this study, the hot gas produced by the electrochemical reaction of the SOFC supplies the hot source of the Stirling Engine. Thereby a part of the heat produced by the fuel cell stack is converted into mechanical work, then electrical work through an alternator.

C. *Interest of hybrid systems based on Solid Oxide Fuel Cell and Stirling Engine*

Cogeneration, also call combined heat and power (CHP), is a way to produce simultaneously electrical energy and heat from fossil resources (oil, coal, natural gas, uranium...) or renewable resources (biomass, solar...).

Exergy of an electrical work is higher than every heat flow exergy, whatever its temperature. The objective of the hybridization of SOFC / Thermal Systems (TS) will be to optimize the electrical global production of the system. In the actual context, where energetic efficiency conversion is necessary, this approach is justified [11].

Different TS are used for cogeneration with SOFC. Different thermodynamics cycles can be considered: preheated and non preheated Rankine, recuperative Brayton and Stirling cycles. The first three are often used in the literature [12] [13] [14]. D. Sanchez shows the theoretical evolution of the efficiency of these systems against reference temperature of the hot source of the TS FIG. 2. The interest to use the Stirling Engine with SOFC is higher than the other systems [12]. The objective of this study is to identify the theoretical and experimental potential of coupling SOFC / SE, in the general context of transportation applications. This first paper concerns nevertheless mostly the model of SOFC for combined cycles.

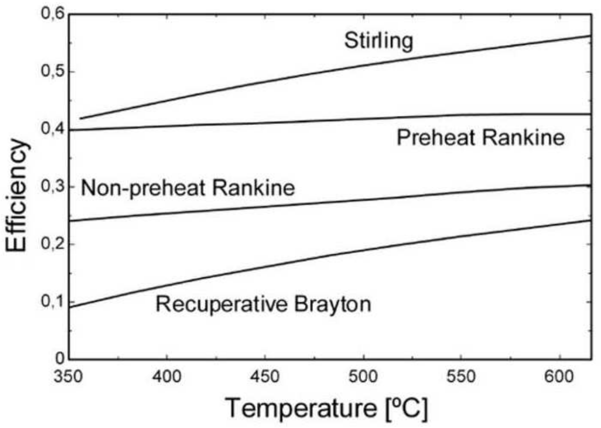

Figure 2: *Efficiency of TS against reference temperature (Fig from [12])*

III. PRESENTATION OF THE ENERGETIC MACROSCOPIC REPRESENTATION

Among methods used for macroscopic modelling approaches of multiphysics phenomena, nodal methods [15] have been widely used. Moreover, bond graph, fuzzy approach can also be cited [16] [17]; these approaches have already been applied to SOFC to improve their dynamical performances [18] [19]. EMR, initially developed to model and control electromechanical systems, has been recently extended to multiphysics systems [3].

A. Fondamentals of EMR

EMR formalism is based on three rules governed by the laws of physics. Noted that they are valid whatever the scale considered in the system (system, subsystem, or component).

Power	**Effort Variable**	**Flow Variable**
Electrical	Voltage [*V*]	Current [*A*]
Mechanical (translation)	Force [*N*]	Velocity [$m.s^{-1}$]
Mechanical (rotation)	Couple [*N.m*]	Angular velocity [$rad.s^{-1}$]
Hydraulic	Pressure [$N.m^{-2}$]	Flow [$m^3.s^{-1}$]
Thermal	Temperature [*T*]	Entropy flow [$W.K^{-1}$]
Chemical	Free Energy [$J.mol^{-1}$]	Molar flow [$mol.s^{-1}$]

Table 1: *Expression of variables effort and flow for different types of power*

The key point of the EMR formalism is the notion of integral causality of magnitudes exchange between the system and its environment. This concept allows a rapid deduction of the control structure by inversion of the blocks. Also the principle of action and reaction must be respected. Through its aspect of immediacy, this concept allows the direct link between power and energy. Finally, the power (energy flow) exchanged by the different subsystems, like bond graph, is written as the product of an effort variable by a flow variable. Table 1 shows the different variables corresponding to each area of physics [4].

B. Elements graphic representation

EMR formalism is based only on few types of elements: source, mono- and multi- physic transformation, and accumulation. The source element allows the representation of the system’s interaction with its environment. The source is considered both as a source- or a well- power. The processing elements contain mono- of multiple physic relations of power conversion. Conversely to the processing elements that represent a direct temporal link between the instantaneous power entering and leaving, the element, the accumulation element represents physical phenomena that cause a temporal phase shift.

The graphical representation of the various icons in EMR formalism is given in Table 2.

C. Control deduction

The control structure of the system is automatically deduced from the EMR model using the principle of inversion. The quantity exchanges between different EMR blocks of the model are explained through the effort and flow variables. Thus the location of the sensors (actual sensors and / or virtual sensors) is directly deducted [3] [4] [5].

IV. EMR FOR SOFC COMBINED HEAT AND POWER: MODEL DESCRIPTION

The representation of the SOFC through EMR formalism is an adaptation of EMR PEMFC model previously published [4]. The modification of the parameters that differentiate the two technologies has been done. Moreover, a global thermal balance of the Fuel Cell stack enabling the ability to evaluate and to locate the different heat flow exchanges between the FC and its environment has also been added. Thus the model can be coupled with other models of heat engines in order to assess the potential for cogeneration.

The model description details the different physical phenomena (Electrochemical, Electrical, Fluidic, and Thermal). The FC supply is provided here with pure hydrogen at the anode and ambient air at the cathode.

A. Fuel Cell Electrochemistry

Fuel cells convert chemical energy directly into electrical and thermal energy. The equation of oxidation of di-hydrogen is given by the chemical equation (4).

This equation is independent of the process itself. Thus the aim of this part is to identify the reversible FC in order to avoid the imperfections (activation, ohmic and concentration losses)

1) Reversible Fuel Cell

The concept of reversible cell is described from the first and second law of thermodynamics. Thus the reversible work produced by the FC is equal to the Gibbs free energy of the reaction (4). The evaluation of the reversible thermodynamic potential and the flow of entropy generated by the reaction in a cell are deduced from the expression of Gibbs [20] [21].

a) Atmospheric pressure thermodynamic reversible potential

$$E_0 = -\frac{\Delta G_0(T_{FC})}{n^{el} \cdot F} = -\frac{\Delta H_0(T_{FC}) - T_{FC} \cdot \Delta S_0(T_{FC})}{n^{el} \cdot F} \qquad (6)$$

Where ΔH_0 and ΔS_0 represent respectively the enthalpy and entropy of the reaction produced in the stack at atmospheric pressure. These quantities are expressed as polynomials functions [22] of the FC temperature (T_{FC}). n^{el} is the number of electrons released during the process of ionization of di-hydrogen (here 2), and F is the Faraday constant.

b) Atmospheric pressure entropy flux generated by the chemical reaction

$$\Delta Sq_0 = \Delta S_0(T_{FC}) \cdot \frac{I_{FC}}{n^{el} \cdot F} \qquad (7)$$

Where I_{FC} is the current supplied by the FC, the other quantities have been defined previously. The relationship between the flow of water produced and the current supplied by the FC (through Faraday's law) is only possible through the hypothesis that the current efficiency is 1.

The two quantities have been defined at atmospheric pressure, thus it is required to make a pressure correction of the FC potential. The variation of the FC potential vs. the partial pressure of the catalytic sites is defined by the Nernst equation [23] [24].

c) Cell Nernst potential

The Nernst potential E_N of the chemical reaction (1) is described by the expression (8):

$$E_N = E_0 + \Delta E \qquad où \qquad \Delta E = \frac{R \cdot T_{FC}}{2 \cdot F} \ln\left(\frac{p_{caH_2} \cdot p_{ccO_2}^{1/2}}{p_{caH_2O}}\right) \qquad (8)$$

Where p_{c*} are the partial pressures of gases (di-hydrogen, di-oxygen, water vapour) on the catalytic sites of the FC (anode, cathode). This potential shift causes an evolution of heat produced by the stack. The heat flow is given by equation (9).

$$\Delta Sq_n = \frac{\Delta E \cdot I_{FC}}{T_{FC}} \qquad (9)$$

The gas consumption on the catalytic sites leads an evolution of gas flows. Equation (10), calculated from Faraday's laws and the ideal gas law, gives the volume flow consumed by the anode and cathode of the FC [2] [5]. n^{el2} represents the number of electrons released in the half-ionization reactions (2 for H_2 and 4 for O_2) [25].

$$q_{Cx} = \frac{I_{FC}}{n^{el2}} \cdot \frac{R \cdot T_{FC}}{p_{Cx}} \qquad (10)$$

Thus, in EMR formalism, the reversible Fuel Cell is represented on Figure 3 by the fluidic / thermal / electrical and electrical / thermal elements conversion.

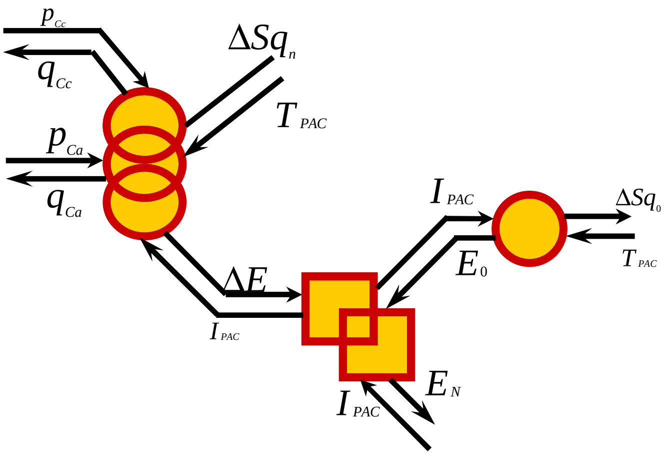

Figure 3: *Reversible potential Fuel Cell EMR [4][5][6]*

Once the reversible potential calculated, the different imperfections of the stack have to be detailed. These sources of irreversibility will generated a potential loss of the FC.

2) Fuel Cell Imperfection

For macroscopic scale modeling approach, three irreversibility sources (also call over-voltage) are identified: activation, ohmic, and concentration over-voltage. Voltage drop and heat generated by theses losses are explained as functions of physical parameters and fuel cell operating conditions.

a) Activation over voltage

The activation losses are due to the electron transfer and to the chemical bond gas molecules modification at the anode and the cathode. A portion of the available energy is consumed by these phenomena. However, the oxidation reaction of di-hydrogen at the anode is faster than the reduction of di-oxygen at the cathode. Therefore, the charge transfer at the cathode will be considered as dominant [4] [26]. The relationship between voltage loss and the FC current is described by the Tafel relationship (11).

$$\Delta V_{act} = \frac{R \cdot T_{FC}}{\alpha \cdot n^{el} \cdot F} \cdot \ln\left(\frac{I_{FC}}{I_0}\right) \qquad (11)$$

Where α is the coefficient of charge transfer, usually given to 0.5 in the literature [13]. I_0 is the exchange current at the interface electrolyte / electrode. These two factors are held constant, and can be determined empirically.

b) Ohmic over voltage

The ohmic losses are represented by a resistance term (R_{ohm}). The latter represents the resistance encountered by the ions and electrons through the various materials constituting the cell stack. The total ohmic resistance can be broken down into four resistances: resistance to the flow of O^{2-} ions through the electrolyte (R_e), R_a and R_c represent the resistance opposing the flow of electrons at the anode and at the cathode, finally a contact resistance due to the different FC components [4]. This resistance will be neglected with respect to the orders of magnitudes of the other three resistors.

$$\Delta V_{ohm} = (R_e + R_a + R_c) \cdot I_{FC} \tag{12}$$

c) Concentration over voltage

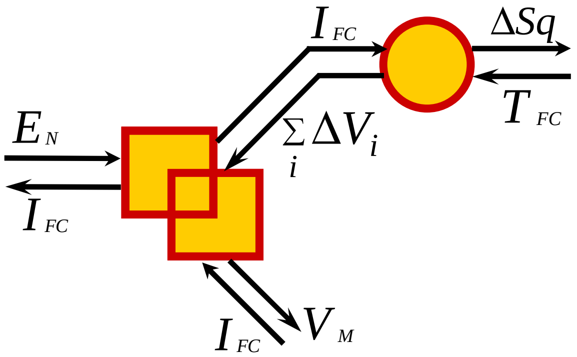

Figure 4: *Fuel Cell Irreversibility EMR [4][5][6]*

The concentration losses are predominant for high current. In these conditions, the reagents are directly consumed by entering the catalytic sites [4]. This therefore leads to a reagent concentration decrease, and thus a voltage drop of the FC.

$$\Delta V_{con} = -\frac{R \cdot T_{FC}}{n^{el} \cdot F} \cdot \ln(1 - \frac{I_{FC}}{I_L}) \tag{13}$$

Where I_L is the current limit, current for which we observe the voltage drop of the FC. As I_0, I_L is determined empirically. Note however that the actual application of such a current during experimental test produces a sharp drop of the FC performance (even leading to its destruction).

Thus we note ΔV the voltage drop due to the over voltage activation, ohmic and concentration losses. It then proceeds to the equation (14). This voltage causes the flow of entropy ΔSq expressed by equation (15).

$$V_M = E_N - \sum_i \Delta V_{irr} \tag{14}$$

$$\Delta S_q = \frac{I_{PAC} \cdot \sum_i \Delta V_{irr}}{T_{FC}} \tag{15}$$

Figure 4 shows graphically according to the EMR formalism, the irreversibilities in one single cell of the FC stack.

B. Fuel Cell fluidic

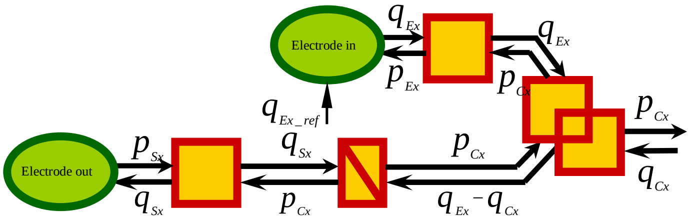

Figure 5: *EMR Fluidic Part[4][5][6]*

This part of the model is the same as the one considered in the PEMFC model [4]. The only changes concern the numerical values of the parameters that are specific to the system. Thus the text of this part contains only the equations to be implemented in the EMR subsystem fluidic model parts. The graphical EMR representation is then shown on figure 5.

$$p_{Ex} = p_{Cx} + R_{dx1} \cdot q_{Ex} \tag{16}$$

$$\frac{dp_{Cx}}{dt} = \frac{1}{C_{dx}} \cdot (q_{Ex} - q_{Cx} - q_{Sx}) \tag{17}$$

$$q_{Sx} = \frac{p_{Sx} - p_{Cx}}{R_{dx2}} \tag{18}$$

C. Fuel Cell thermalbalance

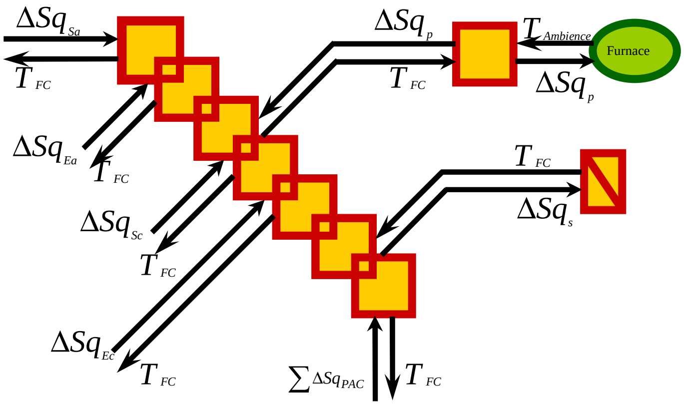

Figure 6: *EMR Thermal Balance of the Fuel Cell*

This part is an evolution of the previous model [4][5][6]. The goal is to identify the thermal potential of the fuel Cell at the anode and the cathode. The scale studied here is macroscopic. The fuel cell is considered as a mass and heat exchanger. The thermal balance of the fuel cell is giving under the relation (19). The EMR representation is shown on figure 6.

The relation (20) provides the balance of heat produced by the chemical reaction and the irreversibility over-voltages. N represents the numbers of cells that composed the stack.

$$\dot{Q}_{ia} + \dot{Q}_{ic} - \dot{Q}_{oa} - \dot{Q}_{oc} - \dot{Q}_{exc} + \dot{Q}_{p} = m_{FC} \cdot Cp_{FC} \cdot \frac{dT}{dt} \tag{19}$$

$$\dot{Q}_{pr} = N \cdot \sum \Delta Sq_{FC} \cdot T_{FC} = N \cdot \left(\Delta Sq_{n} + \Delta Sq_{0} + \Delta Sq\right) \cdot T_{FC} \tag{20}$$

D. *EMR expansion for thermodynamics*

When using the EMR formalism, different areas of physics are studied separately. The distinction is made between the electrical, thermal, fluidic, mechanical and chemical domains to work with effort and flow variables adapted to the formalism. However, in thermodynamics, thermal, fluidic and chemical properties are not separable because of the ideal gas law. Hence there is a request to develop a schematic block well-fitted to our problem in order to keep all informations concerning the gas studied (Figure 7).

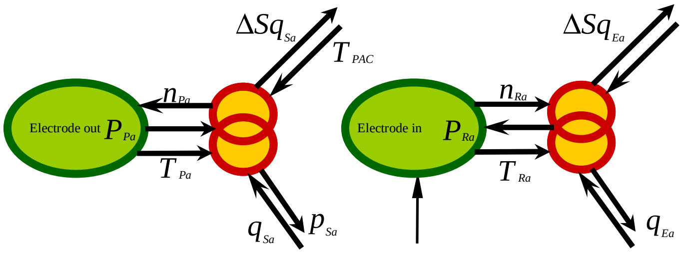

Figure 7 : *EMR thermodynamic coupling gas inlet (right) and outlet (left)*

Thus, for fuel cell applications, the fluidic part of the inlets and the outlets are different through the senses of the variables of action / reaction (flow and pressure) exchanged with the environment. On FC inlet, the gas flow is imposed to the stack to control the combustion. Therefore the pressure is imposed by the FC stack is itself to the incoming gas. On the outlet, the FC stack imposes the outgoing gas flow to the environment, and the pressure is imposed by the environment to the FC.

The calculation of the entropy flow carried by the gas at the inlet and outlet of the stack is achieved by applying the second law of thermodynamics in open systems for stationary flow conditions (valid for the operating conditions used of the FC). The heat flow (21) takes into account the sensible heat of the gases that passes through the FC. The hypothesis is done according to the operating conditions of pressure (around 1bar) and temperature (750-800°C) of SOFC stack.

$$\Delta Sq_{x} = \dot{n}_{x} \cdot M_{x} \cdot Cp \cdot \ln\left(\frac{T_{FC}}{T_{x}}\right) \tag{21}$$

The assembly of the EMR of the various subsystems yields to the overall EMR scheme. This latter can easily be implemented in Matlab Simulink ^{TM} environment to be evaluated.

E. *Global EMR of CHP SOFC*

The overall pattern EMR SOFC for CHP applications is shown in Figure 8.

V. SIMULATION RESULTS

After applying the SOFC physical relations under EMR formalism, the obtained overall model is directly introduced under Matlab Simulink. Figures 8 and 9 establish the comparison between simulation and experimental results of the electrical part of the SOFC system. Experimental results have been performed in our laboratory on a SOFC S-Design 3 cells from HT Ceramix company [25]. According to these results, it can be considered that the electrical part of our model represent well the reality. The thermal validation of the model needs an integration of the thermal balance of the fuel cell environment (Furness). That part will be developed in a further study.

VI. CONCLUSION AND PROSPECTS

EMR formalism based modelling for SOFC Combined Heat and Power showed interesting results. According to this formalism, the identification of causal relationship between the input and output of the systems (or subsystems), the understanding of the interactions mechanisms among the systems (or subsystems), the influences of various system parameters can be easily identified. The following of this work will now consist on the establishment of the MCS (Maximal Control Structure) of the proposed system, in order to define efficient control laws of this multiphysics system and to identify the localization of the actual and of the software sensors. Even more the same modelling approach will be applied to the Stirling Engine, in order to finally make the coupling of the twice systems and to evaluate its overall electrical efficiency.

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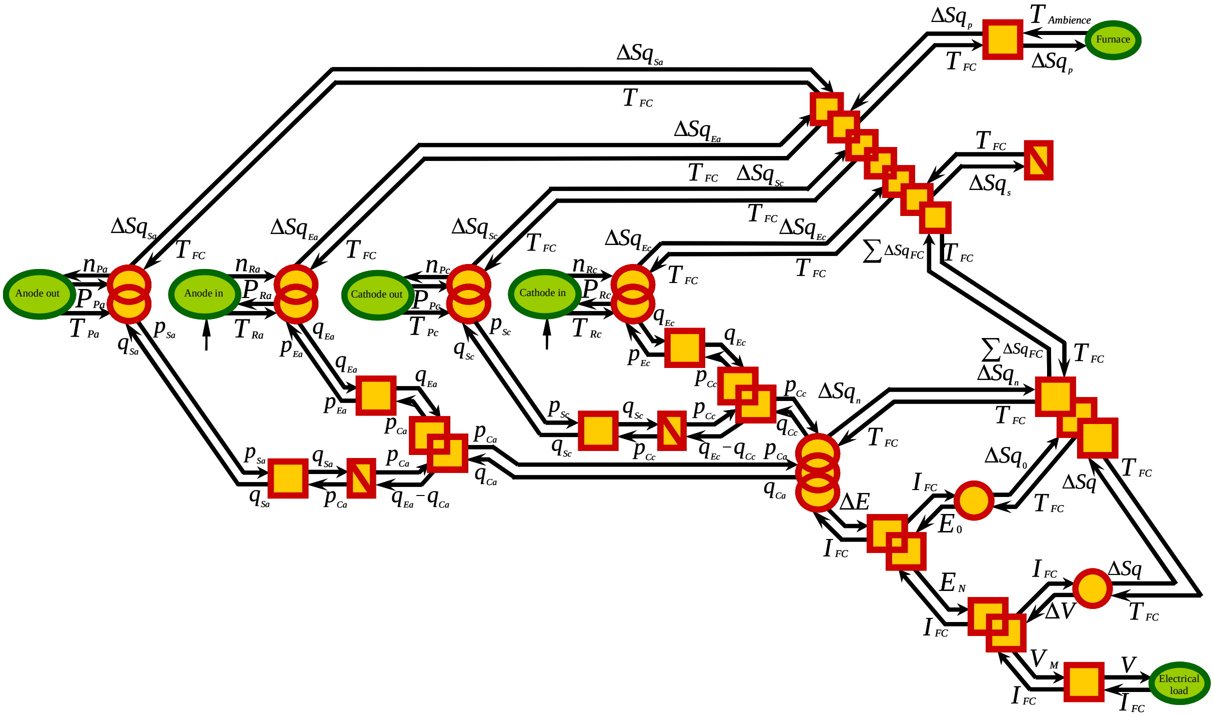
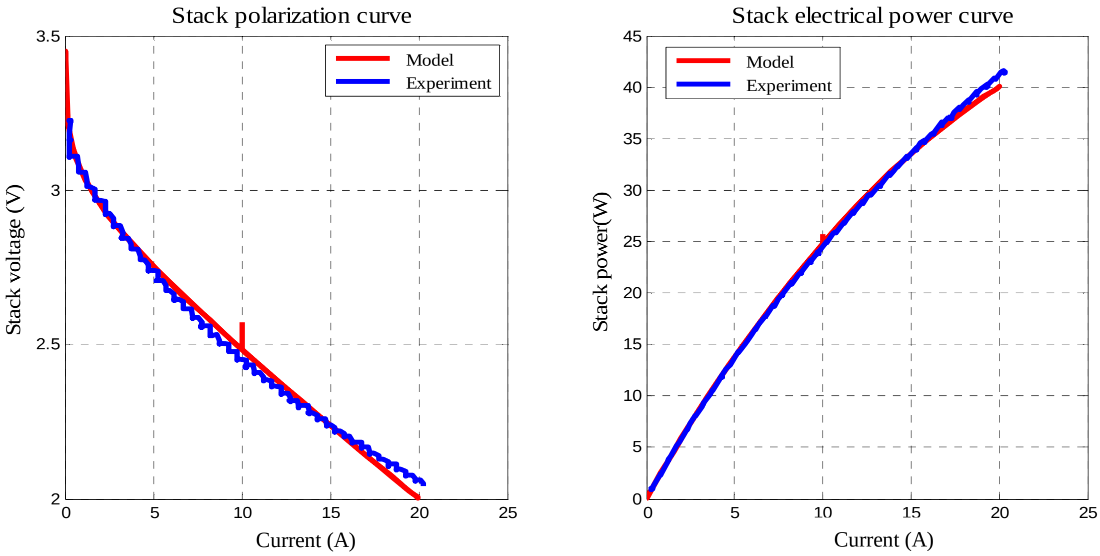


Figure 8: *Global EMR of SOFC for CHP applications*



Figures 9 and 10: Polarization and Power electrical Curves of an *HT Ceramix SOFC stack S-Design 3 Cells*

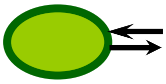
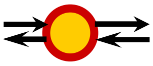
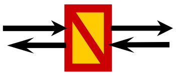
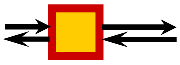
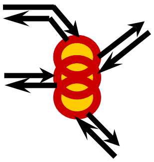
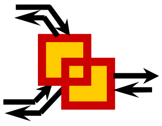
	Source		Multi physic transformation		Accumulation
	Mono physic transformation		multi physic Coupling		mono physic Coupling

Table 2: *Summary of the EMR formalism icons*