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## CONTROL-ORIENTED MODELLING OF FUEL PROCESSING REACTORS IN FUEL CELL POWER SYSTEMS

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## ABSTRACT

Fuel cell represents a new area of interest for power system engineers and researchers. This paper presents control-oriented physics-based modelling of fuel processing system (FPS) reactors in fuel cell power systems. Basically, the FPS converts hydrocarbons to hydrogen rich gas and cleans harmful species according to fuel cell requirements. The physics-based model has importance of understanding the system and useful for model based control designs. The model also can be used for model-based observer designs to estimate unmeasured states or eliminate the sensors.

Keywords: fuel processing, cell power systems, sensors

## **1. INTRODUCTION**

Fuel cell systems represent an alternative power source to traditional, combustion based technologies and batteries that are currently used in a wide variety of applications. Fuel cells are electrochemical devices that are clean, quiet, and efficient. They have no moving parts, and operate continuously as long as fuel is supplied. Therefore, they have excellent reliability and long operating lives. In all cases fuel cell technologies have substantially reduced emissions in comparison with conventional technologies [20]. The U.S. Department of Energy projects that if

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\*The work of the author was supported in part by the National Science Foundation under grants ECS- 0226094, ECS-0238268 and Yildiz Technical University, Turkey. a mere 10% of automobiles nationwide were powered by fuel cells, regulated air pollutants would be cut by one million tons per year and 60 million tons of the greenhouse gas carbon dioxide would be eliminated [2]. On the stationary side, fuel cells are ideal for power generation, either connected to the electric grid to provide supplemental power and backup assurance for critical areas, or installed as a gridindependent generator for on-site service in areas that are inaccessible by power lines. Since fuel cells operate silently, they reduce noise pollution as well as air pollution and the waste heat from a fuel cell can be used to provide hot water or space heating. Fuel cells are the best alternative because they combine higher fuel efficiency with low or no pollution, greater flexibility in installation and operation, quiet operation, low vibration, and potentially lower maintenance and capital costs [1].

Recent developments in polymer electrolyte membrane (PEM) and catalyst technology have dramatically increased the power density of fuel

*Received Date : 25.08.2004 Accepted Date: 14.01.2005*  cells, and made them viable for vehicular and portable power applications, as well as for stationary power plants. A typical fuel cell power system consists of numerous interconnected components, as presented comprehensively in the books [8], [22], [25] and more concisely in the survey papers [10], [11], [12] and [40]. The anode side of the cell stack is fed by the fuel processing system (FPS) which reforms natural gas, gasoline, methanol, or other hydrocarbons into hydrogen. An air blower supplies oxygen in the air to the cathode side. In the cell stack hydrogen from the anode reacts with oxygen from the cathode to generate electricity. Then the produced electricity is conditioned by power electronics circuitry according to power demand of the load, Fig 1.

In earlier studies on PEM Fuel Cells, several mathematical models have been presented. Most of them determine the cell voltage as a function of current density and operating conditions. Springer et al. [42], [43] have made a significant contribution to the understanding of the process occurring in a PEM fuel cell; they have presented an isothermal, one dimensional, steady state model for a complete PEM with a 117 Naffion membrane. Bernardi and Verbrugge [7] have developed a model to study the effects of the transport of gases and water vapor in gas diffusion electrodes on the performance of fuel cells. By using a combination of mechanistic and empirical modeling techniques a parametric model of PEM fuel cell is developed in Amphlett et al. [3]. Many publications pursue water and thermal management modeling: Fuller and Newman [14] have developed a two dimensional membrane assembly to examine water and thermal management of a PEM fuel cell. Nguyen and White [30] have modeled a PEM fuel cell to study effectiveness of various humidification designs. Baschuk and Li [6] have presented a model that includes the effect of variable degree of water flooding in the cathode catalyst layer. Rowe and Li [38] have developed a one dimensional non isothermal model of a PEM fuel cell to investigate the effect of various design and operating conditions on the cell performance, thermal response and water management, and to understand the underlying mechanism. Lee and Lalk [26] have presented a technique in which models can be used to determine the fundamental thermal-physical behavior of a fuel cell stack for any operating design configuration. Modeling and and simulation of the PEM fuel cell is reviewed in Keon et al. [24], where mathematical models of the PEM fuel cell that take the form of either empirical parametric models or steady state models are discussed. A dynamic model that allows predicting the voltage performance during transient and steady state conditions of a PEM fuel cell is presented in Haugstetter [21]. Padulles et al. [31] presents a fuel cell plant model for a power system simulator, and establish in terms of time step and building block connectivity. More recently, Eborn et al. [13] have presented system level models of PEM fuel cells.



**Figure 1.** Components of a fuel cell power system. CPO: Catalytic Partial Oxidation Reactor, WGS: Water Gas Shift Reactor, PROX: Preferential Oxidation Reactor

## 1.1 CONTROL AND OBSERVER PROBLEMS IN FUEL CELL POWER SYSTEMS

Among the most challenging feedback problems in fuel cell power systems is the regulation of hydrogen supplied from the FPS to the anode channel of the cell stack. Insufficient supply causes "starvation" of the cell, which means that the platinum catalyst will start consuming the graphite used in the flow fields, and which can cause immediate and permanent damage [28, 41, 44]. While starvation reduces the life of the cell, excessive hydrogen output from FPS reduces its efficiency, and is also undesirable. A relatively high fuel flow results in a substantial surplus of hydrogen leaving the fuel cell with this large

amount of hydrogen passing to the burner firing the reformer. This results in a relatively high reformer temperature with high conversion efficiency, but with low overall plant efficiency, because of the excess amounts of hydrogen being burned rather than used for the direct conversion to electricity [44]. Another crucial problem in Polymer Electrolyte Membrane (PEM) and Phosphoric Acid (PAFC) fuel cells is to maintain the carbon monoxide level below a critical limit (10ppm for PEM fuel cells [16], [22], [25].) Unfortunately, carbon monoxide is always produced during the reforming reaction and it rapidly de-activates the platinum electro catalyst. preventing hydrogen from reacting in the anode side -a phenomenon called "carbon monoxide poisoning". The result of this is a drop in fuel cell voltage and thus in the system efficiency.

The other severe problem in [34] the temperature of CPO must be at a certain point. Excessive high temperature damages CPO catalyst bed permanently while low temperature slows down the fuel reaction rate [45]. Feedback control of fuel cell power systems has recently started to attract attention. Several control problems for fuel cell-powered electric vehicles are outlined in Powers and Nicastri [33]. Mays et al. [27] have presented dynamic response data, and outlined control design approaches for PEM fuel cell vehicles. Boettner et al. [9] have identified control opportunities for the compressor within the fuel cell system. Control oriented models of fuel cell based vehicles and control tasks for these systems are discussed in Guzzella [19]. Control of the electrical power output and oxidant supply in electrical power generation for a fuel cell powered vehicle is discussed in Mufford et al. [29]. Fuel and reaction control issues are discussed in Gagnon et al. [15] and Sasaki [39]. Pukrushpan et al. [35] have derived a lumped dynamic model of the cell stack, and regulated the net power output by controlling the air supply to the cathode. More recently, a comparison of PI and LQG controls for air supply is presented in Rodatz et al. [37]. In a separate paper Pukrushpan et al. [36] have demonstrated a model-based multivariable control design for the FPS to regulate the temperature of CPO and the mole fraction of hydrogen in the anode.

A major obstacle to implementability of these designs is the absence of reliable measurements of hydrogen partial pressure in the cell stack. Existing sensors are not suitable for use with these controllers because, as discussed [16, 23]. commercially available sensors are not designed to operate in a reformate gas environment. The major complaints are that the sensors that do work to varying degrees of success are too big and costly, and sensors that are potentially low cost are not reliable or do not have the required life time [16]. In some cases neither performance nor cost targets can be met. Observer designs would eliminate the need for sensors, make feedback control applicable, and would be useful for monitoring purposes [4], [5], [18].

Physics-based modelling of the fuel cell power systems is highly important to understand its interactions and performance. Also, for model based control applications and observer designs to estimate unmeasured states physics-based models for fuel cell power systems are essential. In this work, fuel FPS part of the fuel cell power system is modelled. Following section explains fuel processing system (FPS) briefly. Next, control oriented models for FPS reactors are given. In section 4, simulation studies are presented.

# 2. FUEL PROCESSING SYSTEM (FPS)

The FPS reforms hydrocarbons into a hydrogenrich gas, and cleans harmful byproducts according to fuel cell requirements [10], [25], [40]. Among several reformer types, such as steam reforming, autothermal reforming, etc., in this study, the catalytic partial oxidation (CPO) is addressed. In this type of reforming process first starts with cleaning of sulfur from raw hydrocarbon to prevent fuel cell anode electrode poisoning. Then, almost-sulfur free hydrocarbon is converted to hydrogen rich gas by CPO and at the final stages of fuel processing, CO is shifted and oxidized to CO2 with a series of water gas shift (WGS) and preferential oxidation (PROX) reactors. (The latter is also known as selective oxidation). Sulfur and CO requirements are different for each type of fuel cell.



Figure 2. Fuel Processing System block diagram.

## 3. CONTROL-ORIENTED FPS MODEL

#### **3.1 MODELING ASSUMPTIONS**

In these designs, it is assumed that uniform conditions inside the reactors, and employed lumped models. This assumption is reasonable because the exit values of mole fractions are of interest, rather than their distributions inside the reactors. Although distributed phenomena, such as thermal waves and reaction zones, may affect the accuracy of lumped models, this effect can be treated as model uncertainty and addressed with robustness tools. It is also assumed that there is no heat transfer resistance between the catalyst and the gas; that is, the solid and the gas have the same temperature. For applications where this resistance is not negligible, a dynamic model of heat transfer can be incorporated in the design.

## **3.2 CATALYTIC PARTIAL** OXIDATION (CPO) MODEL

The two main reactions in the CPO are Partial Oxidation (POX):  $CH4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2$ 

$$+ /_2 O_2 \rightarrow CO + 2II_2$$

and Full Oxidation (FOX):

$$CH4 + 2O2 \rightarrow CO2 + 2H2O \tag{2}$$

Partial oxidation produces hydrogen for the cell stack, but also generates carbon monoxide which causes the poisoning phenomena in the cell stack. Full oxidation is useful because it supplies additional heat, which facilitates the partial oxidation reaction. The reaction rate expressions for full-and partial-oxidation are given by

$$r_{pox} = s rt \tag{3}$$

$$r_{fox} = (1 - s) r_t$$

where *s* is a selectivity variable [32, 34] which depends on the air-fuel ratio, and rt is the total reaction rate given by the empirical expression

$$r_{t} = k_{g}[O2] \frac{[CH4]}{[CH4] + \varepsilon}$$
(5)

which  $[O_2]$ [CH4]represent in and concentrations of oxygen and methane, respectively, and  $k_g$  and  $\mathcal{E}$  are coefficients available from empirical studies. The first term,  $k_g[O_2]$ , in (5) represents the oxygen mass transfer rate from gas phase to the catalyst. The second term,  $\frac{[CH4]}{[CH4] + \varepsilon}$ , is a smoothing

function that accounts for the situation where methane is the limiting reactant.

Denoting by M the vector of molar holdups of each species; that is,

$$M = (M_{N_2}; M_{CH_4}; M_{CO}; M_{CO_2}; M_{H_2}; M_{H_2O}; M_{O_2})^{\prime}$$
(6)

the dynamic model is obtained from mole balance equations

$$\dot{M} = u_{fuel} + u_{air} - F_{out}^{CPO} + q_1 r_{pox} V + q_2 r_{fox} V$$
(7)

where,  $u_{fuel}$ ,  $u_{air}$ ,  $F_{out}^{CPO}$  (mole/sec) are the fuel, air and exit molar flow vectors respectively, V $(m^3)$  is the reactor volume, and  $q_1$  and  $q_2$  are stoichiometric coefficient vectors:

$$q_{I} = \begin{bmatrix} 0 & -1 & 1 & 0 & 2 & 0 & -1/2 \end{bmatrix}'$$
(8)

$$q_2 = \begin{bmatrix} 0 & -1 & 0 & 1 & 0 & 2 & -2 \end{bmatrix}'$$
(9)

Likewise, from the energy balance principle, the dynamics of temperature *T* are given by

(4)

(1)

 $(mc_{p})\dot{T} = u_{jucl} h(T_{jucl}) + u_{air} h(T_{air}) - F_{out}^{CPO'} h(T) + \Delta H_{par} r_{par} V + \Delta H_{jar} T_{jar} V$ (10)

where *T* is the reaction temperature (K), m(kg)and  $c_p(kJ/kg K)$  are mass and specific heat capacity of the catalyst bed, respectively. The terms  $h(T_{fuel})$ ,  $h(T_{air})$  and h(T) are the ideal gas molar enthalpies for each component at the fuel, air and the exit temperatures.  $\Delta H$  is the heat of reaction at reference temperature.

#### 3.3 WATER GAS SHIFT (WGS) MODEL

The water gas shift reactor contains two inlet streams: Reformate gas from CPO and liquid water from a reservoir. In the reactor *CO* reacts with steam and produces hydrogen and carbon dioxide:

$$CO + H_2O \rightarrow CO_2 + H_2.$$
 (11)

A reaction rate expression for WGS, obtained from the Mass Action Law and the Arrhenius Equation, is

$$r = K_{f} e^{\frac{E_{f}}{RT}} [CO][H_{2}O] - K_{b} e^{\frac{E_{b}}{RT}} [CO_{2}][H_{2}]$$
(12)

where  $K_f$ ,  $K_b$ ,  $E_f$  and  $E_b$  are reaction rate parameters.

Denoting M as in (6), the mole balance equations are

$$\dot{M} = u_{gas} + u_{water} - F_{out}^{WGS} + qrV \tag{13}$$

where,  $u_{gas}$ ,  $u_{water}$ ,  $F_{out}^{WGS}$  are the gas, air and exit molar flow vectors respectively, and

$$q = \begin{bmatrix} 0 & 0 & -1 & 1 & 1 & -1 & 0 \end{bmatrix}'$$
(14)

Likewise, the dynamics of the temperature T is,

$$(mc_{p})\dot{T} = u_{gas}'h(T_{gas}) + u_{water}'h(T_{water}) - F_{out}^{WGS'}h(T) + \Delta HrV$$
. (15)

Note that the same dynamic variables (*M* and *T*) and constants (*V*, *m*,  $c_p$  and  $\Delta H$ ) have been used in CPO, WGS and PROX models. No confusion

should arise, however, because the models are derived separately for each reactor.

#### **3.4 PREFERENTIAL OXIDATION** (PROX) MODEL

The PROX is fed by the reformate gas stream from WGS, and by an air blower. The reactions are the carbon monoxide oxidation:

$$CO + \frac{1}{2} O2 \rightarrow CO2$$
,

(16)

and hydrogen oxidation:

$$H2 + \frac{1}{2}O2 \rightarrow H2O. \tag{17}$$

Suitable reaction rate expressions for them are,

$$r_{CO} = s^{PROX} r_t^{PROX}$$
(18)

$$r_{H_2} = (1 - s^{PROX}) r_t^{PROX}$$
(19)

where  $s^{PROX}$  is a selectivity variable and,

$$r_t^{\text{PROX}} = k[\text{CO}][\text{O2}]^{\frac{1}{2}}$$
 (20)

is the total reaction rate. Then,

$$\dot{M} = u_{gas}^{PROX} + u_{air}^{PROX} - F_{out}^{PROX} + q_1^{PROX} r_{CO}V + q_2^{PROX} r_{H_2}V$$
(21)

where,  $u_{gas}^{PROX}$ ,  $u_{air}^{PROX}$ ,  $F_{out}^{PROX}$  are the gas, air and exit molar flow vectors respectively, and,

$$q_1^{PROX} = \begin{bmatrix} 0 & 0 & -1 & 1 & 0 & 0 & -1/2 \end{bmatrix}^{\prime}$$
 (22)

$$q_2^{PROX} = [0 \ 0 \ 0 \ 0 \ -1 \ 1 \ -1/2]^{\prime}$$
 (23)

Similarly, temperature dynamics are given by

$$(mc_{p})\dot{T} = u_{gss}^{PROX'}h(T_{gss}) + u_{air}^{PROX'}h(T_{air}) - F_{out}^{PROX'}h(T) + \Delta H_{CO}r_{CO}V + \Delta H_{H_{2}}r_{H_{2}}V$$
(24)

#### **4 SIMULATION RESULTS**

Now, simulation results are presented for the control-oriented models in Section 3. The software employed in the simulations is MATLAB SIMULINK. The reference values of the flows are obtained from the calculations in [17] for a power level of 150kW. Figures 3 (a) to (g) show the mole fractions of each species and Figure 3 (h) shows the total outflows of the reactors. Solid lines represent CPO, dashes lines represents WGS, and dotted lines represents PROX reactor models. At t = 5sec inlet gas flow is increased by 20%, at t = 10sec decreased by 30% and at t = 15sec inlet air flow is increased by 20%, at t = 20sec decreased by 30% for CPO. Water supply for WGS is increased by 20% at t =25sec and decreased by 30% at t = 30sec. Air supply for PROX is increased by 20% at t =35sec and decreased by 30% at t = 40sec.

#### **5** CONCLUSIONS

Fuel cell power systems promise to change the face of energy production throughout the world. Efficient operation of them requires advanced feedback control designs. Modelling of the system is highly important for model based control applications and observer designs to estimate unmeasured states. Physics-based model for fuel cell power system is also essential for understanding its interactions and performance. In this study, physics-based control oriented modelling of FPS, which is essential part of the fuel cell power system, is presented and simulations for each reactor in different flow conditions are given.



**Figure 3**. CPO-WGS-PROX Models (Section 3) simulation results: a) Mole fraction of N2, b) Mole fraction of CH4, c) Mole fraction of CO, d) Mole fraction of CO2, e) Mole fraction of H2, f) Mole fraction of H2O, g) Mole fraction of O2, h) Out Flow (mole/sec).

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