Model-Based Analysis for the Control of Molten Carbonate Fuel Cell Systems

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Abstract

A first principles-based, non-linear dynamic model for a Molten Carbonate Fuel Cell system is presented, analysed, and further developed for model-based control applications. Transient validation results, using a laboratory-scale test stack, are provided in support of the cell stack model. Physical arguments and time scale separation are used to derive simpler, reduced-order, model structures useful for control analysis and design. Reduced-order models are derived for the cell stack alone as well as for the cell stack integrated with Balance-of-Plant. The results are compared with the full-order models and used in control system design.

Keywords: Control, Dynamics, Molten Carbonate Fuel Cell, Reduced-Order Model

1 Introduction

High temperature Molten Carbonate Fuel Cell (MCFC) systems represent an approach to using fossil fuels efficiently and affordably for the production of electricity. These types of energy plants benefit from enhanced siting flexibility, attributed to virtually nonexistent NOx, SOx and CO emissions. Emissions of CO2 per kWh of electric power are also reduced due to the higher system efficiencies. MCFC systems are additionally fuel-flexible, allowing operation not only on natural gas but also coal gas, biomass derivatives, and others. Finally, the MCFC is lower in cost relative to other fuel cell types that require more exotic materials.

Overall, an MCFC system is a highly integrated process that couples the dynamics of the fuel cells with Balance-of-Plant components: Thermal Management System, Anode Gas Oxidizer (AGO), and Power Conditioning Unit. As a practical tool for understanding the associated dynamic behaviour of fuel cell systems, dynamic simulation has been well established in this area. For MCFC systems, in particular, there have been several developments in the literature. These primarily include considerations of spatial effects. Such approaches include the three-dimensional stack modelling work of He [1] and its extension, also by He [2], to include a lumped-parameter Balance-of-Plant model. Heidebrecht et al. [3] have developed a transient model that includes spatial effects. This model is formulated in terms of dimensionless parameter groups and includes the effect of the catalytic burner. There has also been recent activity in the area of hybrid fuel cell/gas turbine systems. Roberts et al. [4] have reported on the development of transient models based on one-dimensional or lumped-parameter solutions of fundamental equations applied to MCFC systems coupled with gas turbines.

The modelling approach used in this paper is based entirely on a lumped-parameter formulation of first principles equations. The motivation for the work is drawn from model-based control system analysis and design, where simple dynamic representations of fuel cell systems are preferred. A central advantage is that the state equations can be reduced in number or otherwise simplified, based on physical arguments alone. This has the benefit of a simpler control system design or even a simpler controller.

2 Molten Carbonate Fuel Cell

The type of Molten Carbonate Fuel Cell assumed in this development is the Direct FuelCell (DFC), manufactured by FuelCell Energy, Inc. In this technology, natural gas is internally reformed to hydrogen, partially in an internal reforming unit and partially at the cells. Figure 1 illustrates this two-step approach, which is a combination of Indirect Internal Reforming (IIR) and Direct Internal Reforming (DIR). In the

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3 Fuel Cell Stack Dynamic Model

The fuel cell stack model is presented by first listing the main assumptions. This is followed by a brief description of the approach used in writing the final equation set. Validation results are then presented, which compare the model against a laboratory-scale fuel cell stack for given perturbations in load current. Finally, the development of a reduced-order stack model is then illustrated, where slow dynamics are omitted for the purpose of obtaining a simpler model describing short-term grid transients.

3.1 General Model Assumptions

The underlying assumptions in the fuel cell stack model are that of finite dimensional (lumped-parameter) dynamics, separate reactor models for describing anode and cathode mass inventories, and a single thermal balance for describing the stack hardware temperature. In the respective anode and cathode reactor models, all cells are lumped together with a representative average cell voltage. The anode-side reactor model additionally considers that all RU’s are lumped together with all anodes. In all notation that follows anode will refer to the lumped anode/RU quantities while RU will refer to the inlet quantities. A proper distinction between anode and RU will be used later in the calculation of cell voltage.

3.2 Development of State Equations

Specific assumptions in writing separate mole balances for anode-side and cathode-side reactors and in writing an overall thermal balance are:

(i) Gas mixtures are ideal mixtures.
(ii) Thermodynamic properties follow the ideal gas law.
(iii) Exit stream temperatures equal solid mass temperature.
(iv) Accumulation of gas phase enthalpy is negligible compared to that of solid components.

Under these assumptions, mole balances are written for each gas species, \(i\), within the anode and cathode, respectively:

\[
\frac{dx_{i,a}}{dt} = \frac{RT_A}{P_a V_a} \left[ \sum_{i=1}^{g} \chi_{i,a} \left( x_{i,a}^{in} - x_{i,a}^{in} \right) - \sum_{i=1}^{g} R_{i,a} + R_{i,a} \right] \tag{5}
\]

\[
\frac{dx_{i,c}}{dt} = \frac{RT_A}{P_c V_c} \left[ \sum_{i=1}^{g} \chi_{i,c} \left( x_{i,c}^{in} - x_{i,c}^{in} \right) - \sum_{i=1}^{g} R_{i,c} + R_{i,c} \right] \tag{6}
\]

A single energy conservation equation is written to describe the stack solid temperature:

\[
M_x C_{ps} \frac{dT_s}{dt} = \sum_{i=1}^{g} \chi_{i,a} \left( \bar{h}_{i,a}^{in} - \bar{h}_{i,a}^{in} \right) - \sum_{i=1}^{g} \bar{h}_{i,a} R_{i,a} \tag{7}
\]

where WGS refers to the Water-Gas Shift reaction. The actual performance (voltage and power) of the fuel cell for a specified load current is determined by the chemical reactants and products, as well as cell temperature.
\[ R_i = \sum_{j=1}^{\mu} \nu_{ij} r_j \]

where \( \nu_{ij} \) are the stoichiometric coefficients of species \( i \) in reaction \( j \), and \( r_j \) is the rate of reaction \( j \). In the fuel cell stack model, reaction rates \( r_1, r_2, r_3, \) and \( r_4 \) are assigned, respectively, to anodic, WGS, reforming, and cathodic reactions. The anodic and cathodic reaction rates are equal (\( r_1 = r_4 \)) and proportional to stack current by Faraday's Law of Electrolysis. The reforming reaction rate, \( r_3 \), is assumed to be determined by the reforming reaction kinetics [6]. On the other hand, the WGS reaction is assumed to be at equilibrium. Accordingly, the WGS equilibrium condition is as follows:

\[ K_{\text{eq}} = \frac{x_{\text{CO}_2,a} x_{\text{H}_2,a}}{x_{\text{CO}_2,s} x_{\text{H}_2O,a}} = e^{(E_1 + E_2)/T_s} \tag{8} \]

The equilibrium constraint implies that \( r_3 \), in both Eq. (5) and Eq. (7), cannot be evaluated; however, \( r_2 \), can be eliminated from the equation set [6]. This is done by first writing the total rate of production within the anodes, \( R_{i,a} \), as the sum of the terms in \( r_2 \), and the terms not including \( r_2 \). This is then substituted into Eq. (5) (for each of the remaining anode-side gases) as well as into the energy conservation equation, Eq. (7). By differentiating Eq. (8) with respect to time and manipulating the entire equation set, an explicit formulation can be obtained to describe anode-side gas composition as well as the stack temperature. A similar derivation is not needed to describe the cathode-side gas composition, which follows directly from Eq. (6).

In order to concisely write the final equation set, several defined variables and constants will be used. Where individual gas components are referenced, the following general component order vector will be utilized:

\[ S = [H_2, CH_4, CO, CO_2, H_2O, N_2, O_2] \tag{9} \]

### 3.2.2 Anode-Side Definitions

\[ \begin{bmatrix} \bar{r}_{\text{H}_2,a} \\ \bar{r}_{\text{CH}_4,a} \\ \bar{r}_{\text{CO},a} \\ \bar{r}_{\text{CO}_2,a} \end{bmatrix}, \quad \bar{r}_{\text{H}_2O,a}, \quad \bar{r}_{\text{N}_2,a}, \quad \bar{r}_{\text{O}_2,a} \]

\[ R_a = [-r_1 + 3r_3, -r_3, r_3, r_1 - r_3, 0, 0]' \]

\[ \nu_a = [1, 0, -1, 1, -1, 0, 0]' \]

### 3.2.2 Cathode-Side Definitions

\[ \begin{bmatrix} \bar{r}_{\text{H}_2,c} \\ \bar{r}_{\text{CH}_4,c} \\ \bar{r}_{\text{CO},c} \\ \bar{r}_{\text{CO}_2,c} \end{bmatrix}, \quad \bar{r}_{\text{H}_2O,c}, \quad \bar{r}_{\text{N}_2,c}, \quad \bar{r}_{\text{O}_2,c} \]

\[ R_c = [0, 0, 0, -r_1, 0, 0, -0.25r_1]' \]

### 3.2.3 Supplementary Definitions

\[ \begin{align*}
\bar{r}_{\text{H}_2} &= \begin{bmatrix} \bar{r}_{\text{H}_2,a} \\ \bar{r}_{\text{H}_2,c} \end{bmatrix}, \\
\Delta \bar{r}_a &= \bar{r}_{\text{H}_2,a} - \bar{r}_{\text{H}_2,c}, \\
\Delta \bar{r}_c &= \bar{r}_{\text{H}_2,c} - \bar{r}_{\text{H}_2,c} \\
\alpha_1 &= x_{\text{CO}_2,a} \left( \frac{1}{x_{\text{CO}_2,a} + 1} + \frac{1}{x_{\text{H}_2,a} + 1} + \frac{1}{x_{\text{H}_2O,a}} \right) \\
\alpha_2 &= \frac{-V_a P_a x_{\text{CO}_2,a} E_2 (\bar{r}_{\text{H}_2} - \bar{r}_{\text{H}_2})}{(T_s)^2 R M_C c_p} \\
\alpha_3 &= N_{\text{H}_2}^{\text{in}} + r_1 + 2r_3 \\
\alpha_4 &= \frac{RT_s}{V_a P_a} \\
\alpha_5 &= \frac{RT_s}{V_c P_c} \\
\lambda_1 &= \frac{1}{\alpha_1 + \alpha_2} \\
\lambda_2 &= \frac{E_2 x_{\text{CO}_2,a}}{(T_s)^2 R M_c c_p} \left( N_{\text{H}_2}^{\text{in}} (\Delta \bar{r}_a + \bar{X}_a) + N_{\text{H}_2}^{\text{in}} (\Delta \bar{r}_a + \bar{X}_a) - (\bar{r}_{\text{CO}_2,a} + \bar{r}_{\text{CO}_2,a}) \right) - P_{\text{H}_2} - Q_{\text{H}_2} \tag{10g} \\
\lambda_3 &= \frac{1}{M_{\text{CO}_2}} \left[ \bar{r}_{\text{H}_2O,c} + \bar{r}_{\text{CO}_2,c} - r_4 \right] \tag{10h} \\
\lambda_4 &= \frac{\bar{r}_{\text{N}_2,c}}{\bar{r}_{\text{N}_2,a}} \tag{11a} \\
\lambda_5 &= \frac{\bar{r}_{\text{O}_2,c}}{\bar{r}_{\text{O}_2,a}} \tag{11b} \\
\lambda_6 &= \lambda_4 \lambda_5 \tag{11c} \\
\lambda_7 &= \lambda_6 \lambda_5 \tag{11d} 
\end{align*} \]

Using the above definitions, an explicit differential equation set is written to describe anode gas composition, stack temperature, and cathode gas composition.
\[
\begin{align*}
\alpha_3 \left( x_{CO_2,a} + x_{CO_a} \right) - N_{ru}^{in} \left( x_{CO_2,ru}^{in} + x_{CO,ru}^{in} \right) - r_1 - r_3 \right] - \lambda_1 \dot{c}_2 \\
 x_{H_2,O,a} = \frac{x_{CO_2,a} x_{H_2,a}}{K_{eq} x_{CO_a}} \\
 x_{N_2,a} = 1 - \left( x_{H_2,a} + x_{CH_4,a} + x_{CO_a} + x_{CO_2,a} + x_{H_2,O,a} \right) 
\end{align*}
\]

(11e)  
(11f)

3.2.5 Stack Temperature

\[
\frac{dT_c}{dt} = \lambda_3 
\]

(12)

3.2.6 Cathode Gas Composition

\[
\begin{align*}
\frac{dx_{H_2,O,c}}{dt} &= \alpha_5 \left[ N_{c_2}^{in} \left( x_{H_2,O,c}^{in} - x_{H_2,O,c} \right) + \frac{3 x_{H_2,O,c} r_1}{2} \right] \\
\frac{dx_{CO_2,c}}{dt} &= \alpha_5 \left[ N_{c_2}^{in} \left( x_{CO_2,c}^{in} - x_{CO_2,c} \right) + \frac{3 x_{CO_2,c} r_1}{2} - r_1 \right] \\
\frac{dx_{N_2,c}}{dt} &= \alpha_5 \left[ N_{c_2}^{in} \left( x_{N_2,c}^{in} - x_{N_2,c} \right) + \frac{3 x_{N_2,c} r_1}{2} \right] \\
\end{align*}
\]

(13a)  
(13b)  
(13c)

\[
x_{O_2,c} = 1 - \left( x_{CO_2,c} + x_{H_2,O,c} + x_{N_2,c} \right) 
\]

(13d)

Two additional state equations, derived from the ideal gas law, are used to describe the respective anode and cathode compartment pressures:

3.2.7 Anode Pressure

\[
\frac{dP_{an}}{dt} = \frac{RT_s}{V_a} \left[ N_{ru}^{in} - N_a + r_1 + 2 r_2 + \frac{\lambda_3 V_a P_a}{(T_a)^2 R} \right] 
\]

(14)

3.2.8 Cathode Pressure

\[
\frac{dP_{cn}}{dt} = \frac{RT_s}{V_c} \left[ N_{c_2}^{in} - N_c - \frac{3 r_1}{2} + \frac{\lambda_3 V_c P_c}{(T_c)^2 R} \right] 
\]

(15)

Cell voltage is a function of pressure, temperature, gas composition, and current. This is represented by the following expression relating equilibrium cell voltage, polarization, and ohmic losses:

\[
V_{cell} = V_o - \eta_{act} - \eta_{conc} - iz 
\]

(16)

The Nernst equation relates equilibrium cell voltage to standard potential, temperature, and cell reactant/product partial pressures:

\[
V_o = E_o + \frac{RT}{2F} \ln \left( \frac{P_{H_2,a}^{1/2} P_{CO_2,c}}{P_{H_2,O,a} P_{CO_2,a}} \right) 
\]

(17)

The cell voltage, defined by Eq. (16), is an average value determined by bulk temperature and stack current, as well as the respective anode-side and cathode-side pressures and gas compositions. The total stack voltage is obtained by multiplying by the number of cells.

Single-cell experiments have been used to derive performance models for standard potential, activation polarization loss, concentration polarization loss, and ohmic loss. These models have been correlated to stack current density, average temperature, and average partial pressures of reactants and products. In the fuel cell stack model, the average temperature is taken as the arithmetic average of that of the cathode inlet and cathode exhaust. The average cathode partial pressure corresponds to the arithmetic average of the cathode inlet and cathode exhaust mole fractions, but at the exhaust pressure defined by Eq. (15). Similar considerations apply to the anode; however, the anode inlet mole fractions need to be calculated, since the model only defines input mole fractions as those into the RU and not into the anode (RU and anode have been assumed to be lumped). In order to calculate the anode inlet gas composition, it is assumed that both the WGS reaction and the reforming reaction are at equilibrium, corresponding to the RU inlet temperature and anode exhaust pressure. Eq. (18), below, summarizes the dependencies of the terms in Eq. (16) and Eq. (17) for the single-cell performance model at appropriate average values.

\[
E_o = f_1 (T) 
\]

(18a)

\[
\eta_{act} = f_2 \left( T, i, P_{H_2,a}, P_{CO_2,a}, P_{H_2,O,a}, P_{CO_2,c}, P_{O_2,c} \right) 
\]

(18b)

\[
\eta_{conc} = f_3 (i) 
\]

(18c)

\[
z = f_4 (T) 
\]

(18d)

3.3 Transient Validation of the Stack Dynamic Model

In this section the stack dynamic model is validated by comparing the dynamic responses of the model with that of a laboratory test unit (referred to as plant) for a set of rapid load tests [7]. The test unit was a 20 kW, 30 cell stack with a 7,800 cm² active area. Transient tests occurred over 3,000 hours of operation. Test I consisted of a sudden increase and decrease in load current at 25 kW, under constant fuel and oxidant flows. Test II consisted of a series of sudden load changes at 16 kW, also under constant fuel and oxidant flows. The activity of the reforming catalyst was adjusted (in the model) to match the experimental conversions of methane to hydrogen within the stack. Other process conditions were set up in the model to match those of the test stack.

Figure 2 shows the load current used in Test I, while Figures 3-5 show the results. Test I data is logged over 14.8 minutes, at between 1 and 2 minute intervals. Figure 3 shows the comparison between the model average cell voltage and the voltage of a single cell within the test stack, which is representative of a typical cell voltage transient, but not
Fig. 2 Test I load current.

Fig. 3 Test I model average cell voltage and plant cell 11 voltage.

Fig. 4 Test I model DC power and plant DC power.

Fig. 5 Test I model temperature and plant average temperature.

Fig. 6 Test II load current.

representative of the average cell voltage (voltage is logged at a fast rate in Test I). According to Figure 3, both the model and plant show a fast initial response, followed by a settling of the voltage, corresponding to a settling gas composition. Both model and plant also exhibit a similar temperature-dependent voltage performance, although the effect is more obvious from Figure 4, which compares model and plant power, and which also gives a better indication of the accuracy of the model average cell voltage. Model and plant average temperatures are compared in Figure 5.

Figure 6 shows the load current used in Test II, while Figures 7–9 provide the transient results. Test II data is logged over 350 minutes at between 1 and 100 minute intervals. The model-predicted average cell voltage and the same Cell 11 voltage (sampled over much larger intervals than that of Test I) are shown in Figure 7. In these test results, as well as in those of Figure 8, linear interpolation is used between sampling points. Thus, for comparison, the model results also use
the same sampling and linear interpolation. Figure 8 shows model and plant stack power, while Figure 9 shows model and plant average temperature.

3.4 A Simplified, Reduced-Order Model for Grid Transient Studies

In this section a simplified, reduced-order, model is developed for application to grid transient studies where the desired time frame of interest is of the order of seconds following a disturbance [8]. Here, the complexity of the full-order stack model can be significantly reduced by neglecting slow stack temperature dynamics. In practice, stack pressures lie within a tight range (low backpressure from the Heat Recovery Unit) and differential pressure is tightly controlled. Therefore, a reasonable assumption is to assign anode and cathode pressures to fixed atmospheric values over the operating range. Additionally, the low level of nitrogen present in the fuel gas in practice is assumed to be negligible. These simplifying assumptions are summarized:

(i) Stack temperature is constant – fixed at a nominal value.

(ii) Anode and cathode pressures are constant – fixed at atmospheric pressure.

(iii) There is no nitrogen in the fuel gas.

A reduced-order model can be derived by setting \( \lambda_3 = 0 \) (rate of change of stack temperature is zero) in Eq. (10):

\[
\begin{align*}
N_{r_a}^m (\Delta \mathbf{h}_a \cdot \mathbf{X}_a^m) + N_{c}^m (\Delta \mathbf{h}_c \cdot \mathbf{X}_c^m) - (\mathbf{h}_a \cdot \mathbf{R}_a) - (\mathbf{h}_c \cdot \mathbf{R}_c) \\
+ (\mathbf{h}_a \cdot \mathbf{v}_2) \frac{\lambda_1 \lambda_2}{\alpha_4} \\
+ (\mathbf{h}_c \cdot \mathbf{v}_2) (\alpha_3 x_{CO,2} - N_{r_2,CO,2} - r_3) - P_{dc} - Q_a = 0
\end{align*}
\]

(19)

This is then substituted into Eq. (10g), which is rewritten:

\[
\begin{align*}
\lambda_2 \left[ 1 + \frac{(\mathbf{h}_a \cdot \mathbf{v}_2) \lambda_1 \lambda_2}{\alpha_4 (T_a) M_a C_p} \right] = -\alpha_4 x_{CO,2} \{ \cdots \}
\end{align*}
\]

(20)

where \{ \cdots \} represents the large bracketed term in Eq. (10g). The second term on the left-hand side is approximately zero, due to the high temperature and large mass-specific heat of a single stack or stack array, leading to:

\[
\lambda_2 = -\alpha_4 x_{CO,2} \{ \cdots \}
\]

(21)

Inspection of the terms in the right-hand side bracket suggests the substitution of the anode composition states from Eq. (11). This results in:

\[
\begin{align*}
\lambda_2 = x_{CO,2} \left[ \frac{x_{CO,2} + x_{H_2,2} + x_{H_2O,2}}{x_{CO,2} + x_{H_2,2} + x_{H_2O,2}} \right] \\
+ \lambda_1 \lambda_2 x_{CO,2} \left[ \frac{1}{x_{CO,2}} + \frac{1}{x_{H_2,2}} + \frac{1}{x_{H_2O,2}} \right]
\end{align*}
\]

(22)

In deriving Eq. (22), the following numerically equivalent alternative to Eq. (11e) has been used:
\[
\begin{align*}
\frac{dx_{H_2O,\alpha}}{dt} &= -a_4 \\
\{a_3 \left(x_{H_2O,\alpha} + x_{CO_2,\alpha}\right) - N_{\text{in}}^{\text{in}} \left(x_{H_2O,\text{ru}}^{\text{in}} + x_{CO_2,\text{ru}}^{\text{in}}\right) - r_1 + 2r_3\} + \lambda_1 \lambda_2 \\
\frac{dx_{CO_2,\alpha}}{dt} &= a_5 \left\{ N_{\text{in}}^{\text{in}} \left(x_{CO_2,\alpha}^{\text{in}} - x_{CO_2,\alpha}^{\text{in}}\right) + \frac{3x_{CO_2,\alpha} r_1}{2} - r_1 \right\} \\
\frac{dx_{O_2,\alpha}}{dt} &= a_5 \left\{ N_{\text{in}}^{\text{in}} \left(x_{O_2,\alpha}^{\text{in}} - x_{O_2,\alpha}^{\text{in}}\right) + \frac{3x_{O_2,\alpha} r_1}{2} - r_1 \right\}
\end{align*}
\]
the full-order model and reduced-order model, while the comparison results in Figure 11 show system voltage for both the full-order model and reduced-order model.

According to both figures, the reduced-order model approximates the full-order model well for about 50 seconds after the initial transient. After about 100 seconds the effects of temperature can be seen as the gas composition moves to new equilibrium values corresponding to changing temperature, and the system voltage performance changes as a result.

4 Fuel Cell Balance-of-Plant Dynamic Model

A fuel cell Balance-of-Plant (BOP) model is described for an integrated fuel cell stack, AGO, and cathode booster blower. The model is presented by first reviewing the main assumptions. This is followed by the dynamic equations for the components. The development of a simplified, reduced-order BOP model is then described along with simplifying assumptions, where a 15-state model is reduced to a 3-state model for the prediction of stack temperature and pressures, leading to a simpler associated control system design. Results are then provided, comparing the full-order model with the reduced-order model.

4.1 Balance-of-Plant Dynamic Model

Figure 12 shows a schematic for a fuel cell integrated with an AGO and cathode booster blower. In this layout, which shows the cathode gas preparation subsystem, the anode off-gas is oxidized in a catalytic oxidizer or AGO. The AGO exhaust gas is boosted in pressure before being supplied as an oxidant to the fuel cell cathodes. The AGO air intake provides oxygen for the cathodic reaction and is also manipulated to control stack temperature. The variable-speed driven booster blower is responsible for controlling differential pressure between the anode and cathode. Throughout this section it is assumed that the stack array in Figure 12 is composed of 16 fuel cell stacks [9], as was assumed in Section 3.4.

4.1.1 Anode Gas Oxidizer

The chemical reactions that take place within the AGO are:

\[
\begin{align*}
    H_2 + \frac{1}{2}O_2 &\rightarrow H_2O \\
    CO + \frac{1}{2}O_2 &\rightarrow CO_2 \\
    CH_4 + 2O_2 &\rightarrow CO_2 + 2H_2O
\end{align*}
\]

The same principle assumptions made in Section 3.2 are carried over in deriving a dynamic model for the AGO. In addition to these assumptions, the reactions described by Eq. (31) are considered to be complete and spontaneous and the Minimum Theoretical Air (MTA) required to support oxidation is always available, where the latter assumption is always practically met. Given the assumption of complete and spontaneous oxidation reactions, and using the component ordering of Eq. (9), the reaction rates are written by inspection and used in mole balance equations analogous to Eq. (5). Similarly, an energy conservation equation, analogous to Eq. (7), and a pressure equation, analogous to Eq. (14), are both written. The equation set is listed as follows:

\[
a_1 = \frac{RT_{ox}}{V_{ox}P_{ox}} \\
a_2 = 0.5N_{ox}^{gas}(x_{H_2,ox} + x_{CO,ox}) - N_{ox}^{gas} - N_{ox}^{air} \\
\frac{dx_{CO_2,ox}}{dt} = a_1 \left[ a_2 x_{CO_2,ox} + N_{ox}^{gas} \left( x_{CO_2,ox} + x_{CO_2,ox} + x_{H_2O,ox} + x_{CH_4,ox} \right) \right] \\
\frac{dx_{H_2O,ox}}{dt} = a_1 \left[ a_2 x_{H_2O,ox} + N_{ox}^{gas} \left( x_{H_2O,ox} + x_{CO_2,ox} + x_{CH_4,ox} \right) \right] \\
\frac{dx_{N_2,ox}}{dt} = a_1 \left[ a_2 x_{N_2,ox} + N_{ox}^{gas} x_{N_2,air} \right] \\
x_{O_2,ox} = 1 - \left[ x_{CO_2,ox} + x_{H_2O,ox} + x_{N_2,ox} \right] \\
\frac{dT_{ox}}{dt} = \frac{N_{ox}^{gas}(h_{ox}^{gas} - x_{ox}^{gas}) + N_{ox}^{air}(h_{air}^{gas} - x_{air}^{gas}) - N_{ox}(h_{ox} - h_{ox})}{M_{ox}T_{ox}} \\
\frac{dP_{ox}}{dt} = -a_1 P_{ox} \left[ -a_2 + 2N_{ox}^{gas} x_{CH_4,ox} + N_{ox} - \frac{T_{ox}}{a_1 T_{ox}} \right]
\]

where $h^{gas}_{ox}$, $h_{air}$, and $h_{ox}$ are appropriately defined vectors of molar enthalpy of the anode off-gas, air, and oxidizer exhaust, respectively. Similarly, $x^{gas}_{ox}$, $x_{air}$, and $x_{ox}$ are the anode off-gas, air, and oxidizer compositions, respectively.
The cathode booster blower is described by a static model, where the following correlations have been derived and curve-fitted against manufacturer’s performance data.

\[ S_P = \beta_1 v_R^2 + \beta_2 v_R + \beta_3 \]  
\[ \eta = \theta_1 v_R^2 + \theta_2 v_R^2 + \theta_3 v_R + \theta_4 \]  
\[ P_R = \delta_1 v_R + \delta_2 \]  

(36a) \hspace{1cm} (36b) \hspace{1cm} (36c)

where the coefficients \( \beta_i, \theta_i, \) and \( \delta_i \) are constants. The following relationships describe the blower performance at off-rated speed conditions:

\[ v = K \cdot v_R \]  
\[ SP = K^2 \cdot S_P \]  
\[ P = K^3 \cdot P_R \]  
\[ K = \frac{N}{N_R} \]

(37a) \hspace{1cm} (37b) \hspace{1cm} (37c) \hspace{1cm} (37d)

4.2 A Simplified, Reduced-Order Model for Fuel Cell and Balance-of-Plant

In this section, the fuel cell stack model can be further simplified using physical arguments. The 15-state model for integrated stack-oxidizer-booster blower is sufficient for the prediction of stack temperature and pressures; however, it can be simplified for the purpose of control system design. The 15-state model is reduced to one with 3 states, useful for model-based control of stack differential pressure and stack temperature. Several assumptions are initially made with reference to Figure 12:

(i) Fuel gas composition is fixed.
(ii) Fuel gas temperature is a static function of system load.
(iii) Fuel utilization is manipulated by adjusting fuel gas flow-rate.

The first assumption follows from the fact that the anode gas composition is relatively invariant when fuel utilization is maintained at a fixed setpoint. The second assumption is related to the fact that the setpoint of gas temperature into the RU’s is typically a function of system load. The assumption follows, since control of this temperature is very fast (using a fuel superheater and bypass combination). In practice, manipulating the flow of natural gas, which is not represented in this Balance-of-Plant model, maintains fuel utilization. Thus, fuel utilization is maintained by adjusting the flow of fuel gas (steam and natural gas mixture), according to the third assumption.

First, it is realized that the oxidizer volume is relatively small. As a result, the oxidizer mole fraction differential equations are replaced by algebraic equations. After setting derivatives to zero and using “hat” notation to represent fixed nominal values (Assumption (ii)), then Eq. (33) becomes:

\[ x_{CO_2,ax} = \frac{\left( x_{\text{gas},CO_2,ax} + x_{\text{gas},CO,ax} + x_{\text{gas},CH_4,ax} \right) N_{\text{gas},ax}}{N_{\text{air},ax} + \left[ 1 - 0.5 \left( x_{\text{gas},H_2,ax} + x_{\text{gas},CO,ax} \right) \right] N_{\text{gas},ax}} \]  
\[ x_{H_2O,ax} = \frac{\left( x_{\text{gas},H_2O,ax} + x_{\text{gas},H_2,ax} + 2x_{\text{gas},CH_4,ax} \right) N_{\text{gas},ax}}{N_{\text{air},ax} + \left[ 1 - 0.5 \left( x_{\text{gas},H_2,ax} + x_{\text{gas},CO,ax} \right) \right] N_{\text{gas},ax}} \]  
\[ x_{N_2,ax} = \frac{N_{\text{air},x_{N_2,ax}}}{N_{\text{air},ax} + \left[ 1 - 0.5 \left( x_{\text{gas},H_2,ax} + x_{\text{gas},CO,ax} \right) \right] N_{\text{gas},ax}} \]  
\[ x_{O_2,ax} = 1 - \left( x_{CO_2,ax} + x_{H_2O,ax} + x_{N_2,ax} \right) \]

(38a) \hspace{1cm} (38b) \hspace{1cm} (38c) \hspace{1cm} (38d)

The dynamic response of the cathode gas composition is also relatively fast; hence the mole fraction differential equations, Eq. (13), are similarly replaced by algebraic equations:

\[ x_{H_2O,c} = \frac{N_{\text{gas},x_{H_2O,c}}}{N_{\text{gas}} - 3r_1/2} \]  
\[ x_{CO_2,c} = \frac{N_{\text{gas},x_{CO_2,c}}}{N_{\text{gas}} - 3r_1/2} \]  
\[ x_{N_2,c} = \frac{N_{\text{gas},x_{N_2,c}}}{N_{\text{gas}} - 3r_1/2} \]  
\[ x_{O_2,c} = 1 - \left( x_{CO_2,c} + x_{H_2O,c} + x_{N_2,c} \right) \]

(39a) \hspace{1cm} (39b) \hspace{1cm} (39c) \hspace{1cm} (39d)

By assumption, the molar flow of gas into the RU is calculated so that fuel utilization is properly maintained, whereby the flow of gas into the RU is proportional to current, \( N_{\text{gas}} = k_1 I_{sys} \). Faraday’s Law provides an additional relationship, which is employed:

\[ r_1 = k_2 I_{sys} \]

(40)

As far as stack temperature and pressures are concerned, there is little difference in assuming that the reforming reaction is complete and spontaneous. However, under this assumption, the reaction rate expression becomes much simpler:

\[ r_3 = N_{\text{gas}} x_{\text{gas},CH_4} = k_3 I_{sys} \]

(41)

From Eq. (40) and Eq. (41), the parameter \( \alpha_3 \), defined in Eq. (10c), can now be evaluated:

\[ \alpha_3 = \frac{N_{\text{gas}} + r_1}{r_3} = k_4 I_{sys} \]

(42)

The next step in the derivation is to simplify the expressions for ideal gas partial molar enthalpies. All temperatures within the stack-oxidizer-booster blower system fall within a small enough range (for both steady-state and transient) so
that a linear approximation can be used around an appropriate nominal temperature:

$$\dot{h}_i(T) = a_i T + b_i$$  \hspace{1cm} (43)$$

where

$$A = \begin{bmatrix} a_1 \\ \vdots \\ a_d \end{bmatrix}, \quad B = \begin{bmatrix} b_1 \\ \vdots \\ b_d \end{bmatrix}$$  \hspace{1cm} (44)$$

Previous definitions can now be rewritten:

$$\dot{h}_{\text{in}}^{\text{ru}} = AT_{\text{ru}} + B$$

$$\dot{h}_{\text{s}} = AT_{\text{s}} + B$$

$$\Delta \dot{h}_{\text{a}} = \dot{h}_{\text{in}}^{\text{ru}} - \dot{h}_{\text{s}} = A(T_{\text{ru}} - T_{\text{s}})$$

$$\Delta \dot{h}_{\text{a}} \cdot X^{\text{in}}_{\text{ru}} = A \cdot X^{\text{in}}_{\text{ru}} (T_{\text{ru}} - T_{\text{s}}) = k_g (T_{\text{ru}} - T_{\text{s}})$$

$$\Delta \dot{h}_{\text{c}} = \dot{h}_{\text{c}}^{\text{in}} - \dot{h}_{\text{s}} = A(T_{\text{c}} - T_{\text{s}})$$

$$\Delta \dot{h}_{\text{c}} \cdot X^{\text{in}}_{\text{c}} = A \cdot X^{\text{in}}_{\text{c}} (T_{\text{c}} - T_{\text{s}})$$

where $X^{\text{in}}_{\text{ru}}$ is obtained from the fixed inlet composition and $X^{\text{in}}_{\text{c}}$ is left as a free variable, defined by the following:

$$X^{\text{in}}_{\text{c}} = \begin{bmatrix} x^{\text{in}}_{\text{CO}_2} & 0 & x^{\text{in}}_{\text{H}_2} & x^{\text{in}}_{\text{O}_2} & x^{\text{in}}_{\text{H}_2\text{O}} \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix}$$  \hspace{1cm} (45)$$

Additional terms in the stack model are evaluated:

$$\dot{h}_{\text{s}} \cdot R_{\text{s}} = (AT_{\text{s}} + B) \cdot R_{\text{s}}$$

$$= (A \cdot R_{\text{s}})T_{\text{s}} + B \cdot R_{\text{s}} = (k_5 T_{\text{s}} + k_6)I_{\text{sys}}$$  \hspace{1cm} (46)$$

$$\dot{h}_{\text{s}} \cdot R_{\text{c}} = (AT_{\text{c}} + B) \cdot R_{\text{c}}$$

$$= (A \cdot R_{\text{c}})T_{\text{s}} + B \cdot R_{\text{c}} = (k_5 T_{\text{s}} + k_6)I_{\text{sys}}$$  \hspace{1cm} (47)$$

Under the assumption that the anode gas composition is constant, then $\lambda_1 \lambda_2 = 0$, according to Eq. (11c). This simplifies $\lambda_3$ in Eq. (10h). Evaluation of $\lambda_3$, using the above, results in:

$$\lambda_3 = \frac{k_5^2 T_{\text{ru}}^2 - k_11 T_{\text{s}} + k_{12}}{M_{\text{c}} C_{\text{ps}}} I_{\text{sys}} + N^{\text{in}}_{\text{c}} \left( A \cdot X^{\text{in}}_{\text{c}} \right) (T_{\text{s}} - T_{\text{c}}) - P_{\text{dc}} - Q_{\text{a}}$$  \hspace{1cm} (48)$$

The next step is to assume that the oxidizer temperature response is relatively fast. In fact, the total solid mass-specific heat of the oxidizer is at least an order of magnitude less than that of the equivalent stack. Thus, Eq. (34) is replaced by an algebraic equation:

$$\dot{T}_{\text{ox}} = \frac{\left( A \cdot X^{\text{gas}}_{\text{ox}} \right) N^{\text{gas}}_{\text{ox}} T_{\text{gas}} + (B \cdot X^{\text{gas}}_{\text{ox}}) N^{\text{gas}}_{\text{ox}} + (\dot{h}_{\text{ao}} \cdot X_{\text{air}}) N^{\text{air}}_{\text{ox}} - (B \cdot X^{\text{ox}}_{\text{ox}}) N_{\text{ox}}}{(A \cdot X^{\text{ox}}_{\text{ox}}) N_{\text{ox}}}$$  \hspace{1cm} (49)$$

where $\dot{h}_{\text{ao}} = AT_{\text{ao}} + B$. The evaluation of $T_{\text{in}}^{\text{in}}$ in Eq. (48) and $T_{\text{gas}}^{\text{gas}}$ in Eq. (49) follows, where piping heat losses are included:

$$T_{\text{in}}^{\text{in}} = \frac{\left( A \cdot X^{\text{in}}_{\text{c}} \right) T_{\text{blu}} N_{\text{ox}} - Q_1}{\left( A \cdot X^{\text{in}}_{\text{c}} \right) N_{\text{ox}}}$$  \hspace{1cm} (50)$$

$$T_{\text{gas}}^{\text{gas}} = \frac{\left( A \cdot X^{\text{gas}}_{\text{ox}} \right) T_{\text{a}} N_{\text{ox}}^{\text{gas}} - Q_2}{\left( A \cdot X^{\text{gas}}_{\text{ox}} \right) N_{\text{ox}}^{\text{gas}}}$$  \hspace{1cm} (51)$$

Finally, the evaluation of $T_{\text{blu}}$ in Eq. (50) follows from Eqs. (36) and (37) and from standard equations for adiabatic compression. Eq. (35), describing oxidizer pressure, is also reduced to an algebraic equation due to the relatively small volume.

A 15-state dynamic model for an integrated fuel cell stack, Anode Gas Oxidizer, and cathode booster blower has been
reduced to a 3-state model, which retains dynamic states for stack temperature, anode pressure, and cathode pressure. One loop controller is designed for stack temperature, whereas the manipulated variable is oxidizer airflow, while another loop controller is designed for stack differential pressure, where the manipulated variable is booster blower speed. The design for each of the controllers is based on the reduced 3-state model. A comparison is made between the controlled 15-state model and the controlled 3-state model, by simulating a sudden increase in power. The comparison results are shown in Figure (13) for stack temperature and in Figure (14) for differential pressure.

5 Conclusions

The lumped-parameter fuel cell stack dynamic model, based on first principles, is proven to be a viable tool for model-based analysis and control of MCFC systems. The model validates well when compared to laboratory-scale stack test results. Physical arguments can be used to reduce and simplify the equation set for the purpose of either effecting a simpler control design process or generating lower-order controllers, based on lower-order plant models. Extension of the modeling process to include the fuel cell Balance-of-Plant has also resulted in a physically intuitive model that can be simplified. A 15-state model has been reduced to a 3-state model via directions suggested by physical arguments and practical operation. A comparison has been made to justify the types of assumptions made. The reduced-order models will be used in the eventual design of model-based robust controllers for MCFC systems.

List of Symbols

- $E_g$: Standard potential / V
- $E_i$: WGS reaction Gibbs’s free energy constant / 1
- $F$: Faraday’s constant / C mol⁻¹
- $f_{i/n}_{i}$: Partial molar enthalpies at stack temperature / J mol⁻¹
- $h_{i}$: Partial molar enthalpies at stack temperature / J mol⁻¹
- $f_{i/gal_{i}}$: Anode off-gas partial molar enthalpies / J mol⁻¹
- $h_{i}$: Air partial molar enthalpies / J mol⁻¹
- $i$: Cell current density / A cm⁻²
- $I_{eq}$: Stack array system current / A
- $K_{eq}$: WGS reaction equilibrium constant / 1
- $M_{t_{i}}C_{pt}$: Stack solid mass-specific heat product / J K⁻¹
- $M_{t_{i}}C_{px}$: Oxidizer solid mass-specific heat product / J K⁻¹
- $N_{t_{i}}$ (N_{in}$): Total molar flow into RU (cathode) / mol s⁻¹
- $N_{t_{i}}$ (N_{in}$): Molar flow of anode off-gas into oxidizer / mol s⁻¹
- $N_{t_{i}}$ (N_{in}$): Molar flow of air into oxidizer / mol s⁻¹
- $N_{t_{i}}$: Total anode (cathode) exhaust molar flow / mol s⁻¹
- $N$: Actual booster blower speed / RPM
- $N_{R}$: Rated booster blower speed / RPM
- $P_{t_{i}}$: Anode (cathode) exhaust pressures / Pa
- $P_{i}$ (P_{i}$): Anode (cathode) species i partial pressure / 1
- $P_{ox}$: Oxidizer pressure / Pa
- $P_{dc}$: Stack DC power / kW
- $P$: Power delivered to blower / kW
- $Q_{a}$: Stack heat loss / kW
- $Q_{1}$: Piping heat loss (booster to cathode) / kW
- $Q_{2}$: Piping heat loss (AG0 to booster) / kW
- $r_{i}$: Electrochemical reaction rate / s⁻¹
- $R_{i}$: Reforming reaction rate / s⁻¹
- $R_{i,eq}$ (R_{i,eq}$): Total production rate of species i (from all reactions) / mol s⁻¹
- $R$: Universal gas constant / J mol⁻¹ K⁻¹
- $S_{P_{K}}$: Static pressure at rated blower speed / Pa
- $S_{P}$: Static pressure / Pa
- $T_{s}$: Stack solid average temperature / K
- $T_{ox}$: Oxidizer temperature / K
- $V_{e}$: Equilibrium potential / V
- $V_{cell}$: Cell voltage / V
- $V_{o}$: Anode (cathode) compartment volumes / m³
- $V_{ox}$: Oxidizer volume / m³
- $x_{i}$ (x_{i}$): Anode (cathode) exhaust mole fractions / 1
- $x_{i,ox}$ (x_{i,ox}$): RU (cathode) inlet mole fractions / 1
- $x_{i,air}$: Oxidizer exhaust mole fractions / 1
- $z$: Air mole fraction / 1
- $z$: Air mole fraction / 1
- $\eta_{act}$: Cell ohmic impedance / Ω cm
- $\eta_{conc}$: Activation polarization / V
- $\eta$: Concentration polarization / V
- $\rho$: Blower efficiency / 1
- $\rho$: Fluid velocity / m³ s⁻¹
- $\rho_{g}$: Fluid velocity at rated blower speed / m³ s⁻¹
- $\xi$: Total number of gas species / 1

References