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Optimal Load Changes for a Molten Carbonate Fuel Cell Model

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Abstract Molten carbonate fuel cells are a promising technology for future stationary power plants. In order to enhance service life a more detailed understanding of the dynamical behavior is needed. This is enabled by a hierarchy of mathematical models based on chemical and physical laws. These mathematical models allow numerical simulation and optimal control of load changes. Mathematically speaking, we solve an optimal control problem subject to a degenerated partial differential equation system coupled with an integro differential-algebraic equation system. New numerical results are presented.

1 Introduction

Several types of fuel cells exist which are suited for different applications due to their different behavior [7]. Stacks of molten carbonate fuel cells are used for stationary power and heat supply [6]. Due to the high operation temperature an internal reforming, i.e. production of H_2 from CH_4 (or other fuel gases) in the fuel cell, is possible. Moreover clean exhaust gases are produced. In order to enhance service life, hot spots and high temperature gradients inside the fuel cell have to be avoided. Recently a hierarchy of mathematical models for a single (averaged) MCFC has been developed in order to describe the dynamical behavior of important physical and chemical variables [2, 6]. We will demonstrate, how one of these models can

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be used for simulation and optimal control of load changes during electrical power production.

Mathematically speaking, we have to compute optimal boundary control functions for an optimal control problem subject to a partial differential-algebraic equation system coupled with an integro differential-algebraic equation system. Due to the continual model updates during the project time, we chose the approach "first discretize, then optimize". We present recent numerical results for faster load changes [4].

2 Mathematical Model of MCFC

The following 2D crossflow model of a single molten carbonate fuel cell is based on [2, 3], see also [4]. The important mathematical variables are depicted in Fig. 1, which describe the gas flow through the anode gas channel, the catalytic combustor, the reversal chamber, the cathode gas channel and the cathode recycle.

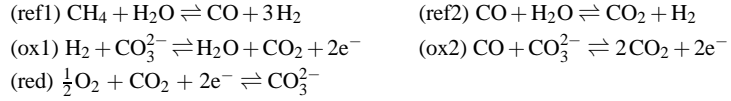
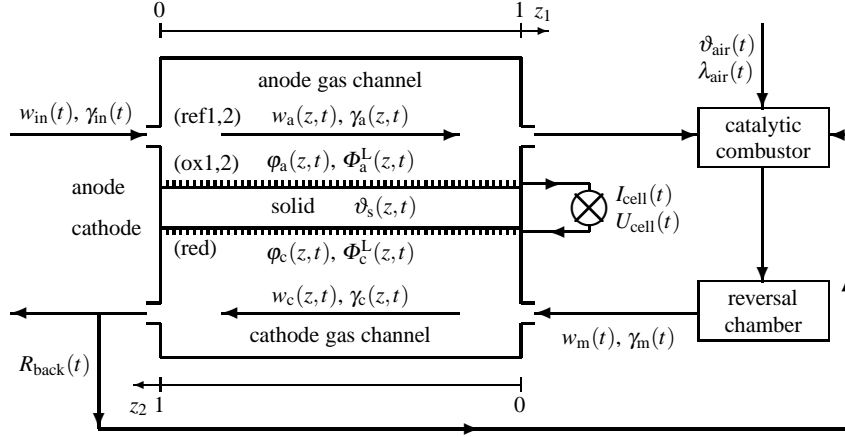


Fig. 1 2D crossflow model of a molten carbonate fuel cell with mathematical variables $w = (\chi, \vartheta)$, χ = vector of molar fractions, ϑ = temperature, φ = vector of partial pressures, Φ = electrical potential, U_{cell} = cell voltage, I_{cell} = cell current

The spatial domain and boundaries are given in Fig. 2. All variables are dimensionless. One unit of the dimensionless time t equals 12.5 seconds. The index set

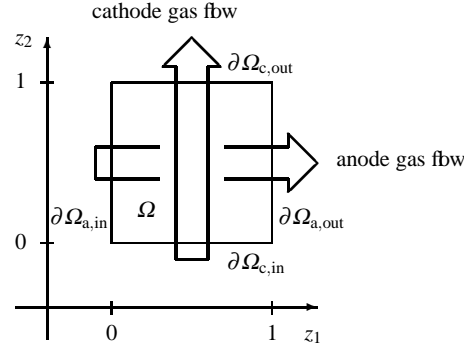


Fig. 2 2D cross flow model: (spatial) domain, boundaries ($\partial\Omega = \partial\Omega_{a,in} \cup \partial\Omega_{a,out} \cup \partial\Omega_{c,in} \cup \partial\Omega_{c,out}$), gas fbw directions

$\mathcal{S} := \{\text{CH}_4, \text{H}_2\text{O}, \text{H}_2, \text{CO}, \text{CO}_2, \text{O}_2, \text{N}_2\}$ is used for molar fractions and partial pressures. In the following there always holds $i \in \mathcal{S}$, $j \in \{a, c\}$.

States, prescribed by PDEs, PDAEs and DAEs:

- In the anode gas channel ($j=a$) resp. the cathode gas channel ($j=c$): molar fractions $\chi_{i,j}(z,t)$, gas temperatures $\vartheta_j(z,t)$, and molar flow densities $\gamma_j(z,t)$. Near the electrodes: partial pressures $\phi_{i,j}(z,t)$. Only a subset of chemical substances is needed during the numerical solution of the mathematical model.
Abbreviation: $w_j(z,t) = ((\chi_{i,j})_{i \in \mathcal{S}}, \vartheta_j)$, $w_{a/c} = (w_a, w_c)$.
- In the solid (electrolyte): temperature $\vartheta_s(z,t)$.
- At the entry of the cathode gas channel: molar fractions $\chi_{i,m}(t)$, temperatures $\vartheta_m(t)$, and molar flow density $\gamma_m(t)$.
Abbreviation: $w_m(t) = ((\chi_{i,m})_{i \in \mathcal{S}}, \vartheta_m)$
- Potentials $\Phi_a^L(z,t)$, $\Phi_c^L(z,t)$, cell voltage $U_{\text{cell}}(t)$.
Abbreviation: $\Phi_{a/c}^L = (\Phi_a^L, \Phi_c^L)$.

Variables, which may serve as components of the boundary control $u(t)$:

- At the entry of the anode gas channel: molar fractions $\chi_{i,in}(t)$, gas temperature $\vartheta_{in}(t)$, and molar flow density $\gamma_{in}(t)$.
Abbreviation: $w_{in}(t) = ((\chi_{i,in})_{i \in \mathcal{S}}, \vartheta_{in})$
- At the entry of the catalytic combustor: Gas temperature $\vartheta_{\text{air}}(t)$, air number $\lambda_{\text{air}}(t)$.
- Switch for cathode recycle: $R_{\text{back}}(t) \in [0, 1]$.

Variables, which are prescribed:

- The cell current $I_{\text{cell}}(t)$ is prescribed, and usually either constant or a step function for a load change from one constant level to another constant level.

Partial differential-algebraic equations with boundary conditions:

$$\frac{\partial \vartheta_s}{\partial t} = \mu_1 \frac{\partial^2 \vartheta_s}{\partial z_1^2} + \mu_2 \frac{\partial^2 \vartheta_s}{\partial z_2^2} + \psi_1(\vartheta_s, w_{a/c}, \varphi_{a/c}, \Phi_{a/c}^L, U_{\text{cell}}), \quad \frac{\partial \vartheta_s}{\partial z} \Big|_{\partial \Omega} = 0, \quad (1)$$

$$\frac{\partial w_a}{\partial t} = -\gamma_a \vartheta_a \frac{\partial w_a}{\partial z_1} + \psi_2(\vartheta_s, w_a, \varphi_a, \Phi_a^L), \quad w_a \Big|_{\partial \Omega_{a,\text{in}}} = w_{\text{in}}(t), \quad (2)$$

$$\frac{\partial w_c}{\partial t} = -\gamma_c \vartheta_c \frac{\partial w_c}{\partial z_2} + \psi_3(\vartheta_s, w_c, \varphi_c, \Phi_c^L, U_{\text{cell}}), \quad w_c \Big|_{\partial \Omega_{c,\text{in}}} = w_{\text{m}}(t), \quad (3)$$

$$0 = -\frac{\partial(\gamma_a \vartheta_a)}{\partial z_1} + \psi_4(\vartheta_s, w_a, \varphi_a, \Phi_a^L), \quad \gamma_a \Big|_{\partial \Omega_{a,\text{in}}} = \gamma_{\text{in}}(t), \quad (4)$$

$$0 = -\frac{\partial(\gamma_c \vartheta_c)}{\partial z_2} + \psi_5(\vartheta_s, w_c, \varphi_c, \Phi_c^L, U_{\text{cell}}), \quad \gamma_c \Big|_{\partial \Omega_{c,\text{in}}} = \gamma_{\text{m}}(t), \quad (5)$$

$$0 = \psi_6(\vartheta_s, \chi_a, \varphi_a, \Phi_a^L), \quad 0 = \psi_7(\vartheta_s, \chi_c, \varphi_c, \Phi_c^L, U_{\text{cell}}), \quad (6)$$

$$\frac{\partial \Phi_{a/c}^L}{\partial t} = \psi_8(\vartheta_s, \varphi_{a/c}, \Phi_{a/c}^L, U_{\text{cell}}, I_{a/e/c}; I_{\text{cell}}). \quad (7)$$

Integro differential-algebraic equations:

$$\frac{dU_{\text{cell}}}{dt} = \psi_9(I_{a/e/c}; I_{\text{cell}}), \quad I_a(t) = \int_{\Omega} i_a(\vartheta_s, w_a, \varphi_a, \Phi_a^L) dz, \quad (8)$$

$$I_c(t) = \int_{\Omega} i_c(\vartheta_s, w_c, \varphi_c, \Phi_c^L, U_{\text{cell}}) dz, \quad I_e(t) = \int_{\Omega} i_e(\Phi_{a/c}^L) dz, \quad (9)$$

$$\frac{dw_{\text{m}}}{dt} = \psi_{10}(w_{\text{m}}, \int_{\partial \Omega_{a,\text{out}}} w_a dz_2, \int_{\partial \Omega_{a,\text{out}}} \gamma_a dz_2, \int_{\partial \Omega_{c,\text{out}}} w_c dz_1, \int_{\partial \Omega_{c,\text{out}}} \gamma_c dz_1, \lambda_{\text{air}}, \vartheta_{\text{air}}, R_{\text{back}}), \quad (10)$$

$$\gamma_{\text{m}}(t) = \psi_{11}(w_{\text{m}}, \int_{\partial \Omega_{a,\text{out}}} w_a dz_2, \int_{\partial \Omega_{a,\text{out}}} \gamma_a dz_2, \int_{\partial \Omega_{c,\text{out}}} w_c dz_1, \int_{\partial \Omega_{c,\text{out}}} \gamma_c dz_1, \lambda_{\text{air}}, \vartheta_{\text{air}}, R_{\text{back}}). \quad (11)$$

Initial conditions:

$$\begin{aligned} \vartheta_s \Big|_{t=0} &= \vartheta_{0,s}(z), \quad w_a \Big|_{t=0} = w_{0,a}(z), \quad w_c \Big|_{t=0} = w_{0,c}(z), \quad w_{\text{m}} \Big|_{t=0} = w_{0,\text{m}}, \\ \Phi_{a/c}^L \Big|_{t=0} &= \Phi_{0,a/c}^L(z), \quad \Phi_c^L \Big|_{t=0} = \Phi_{0,c}^L(z), \quad U_{\text{cell}} \Big|_{t=0} = U_{0,\text{cell}} \end{aligned} \quad (12)$$

The PDAE system consists of a parabolic heat equation (1), hyperbolic transport equations (2–3) with given wind direction (because it is known a priori that $\gamma_{a/c}, \vartheta_{a/c}$ are positive), and partial differential-algebraic equations (4–7). A detailed index analysis of (1–12) yields differential time index $\nu_t = 1$ (see [5], some small modifications have to be made in order to allow a switched on cathode recycle). Therefore *consistent* initial conditions are given by (12), no initial conditions can be prescribed for the algebraic variables $\gamma_{a/c/m}, \varphi_{a/c}, I_{a/c/e}$.

An obvious engineering approach is to use the method of lines (MOL) by discretizing the spatial partial derivatives. This is simplified by the a priori knowledge of the wind direction of the hyperbolic equations. A five-point star for the (scaled) Laplacian and suitable upwind formulas are used for the spatial partial derivatives and quadrature formulas for the spatial integrals. This yields a very large semi-explicit DAE of index $\nu = 1$.

$$M\dot{x}(t) = g(x(t), u(t)), \quad M[x(0) - x_0] = 0, \quad M = \text{diag}(I, O) \quad (13)$$

3 Optimization Scenarios and Numerical Results

One drawback of molten carbonate fuel cells is the slow system reaction for a load change (i.e. a change in the cell current I_{cell} during the operation of the MCFC). The potentials $\Phi_{\text{a/c}}^L$, U_{cell} react very fast, whereas the molar quantities and especially the solid temperature ϑ_s react only slowly. Fast load changes induce temperature changes, which have to be compensated especially for increasing cell current.

The goal of the optimal control is therefore to control the system after a load change as fast as possible into the new stationary state. A good indicator for the stationary state is the cell voltage U_{cell} , which reacts very fast and significantly on a sudden load change of the cell current and moreover reacts also on changes of the slowest variable, the solid temperature ϑ_s .

The following technologically interesting scenario is analyzed [4]: The system input cell current is prescribed as a discontinuous step function

$$I_{\text{cell}}(t) = \begin{cases} I_{\text{cell},1} = 0.7 & \text{if } t \leq t^* \\ I_{\text{cell},2} = 0.6 & \text{if } t > t^* \end{cases} \quad (14)$$

Initial conditions (12) at $t = 0 \leq t^*$ are the stationary solution for constant $I_{\text{cell},1}$.

Find optimal boundary control functions $u : [0, t_f] \rightarrow \mathbb{R}^6$, such that the functional

$$J[u] = \int_{t^*}^{t_f} L dt \text{ with } L = [U_{\text{cell}}(t) - U_{\text{cell},2,\text{stat}}]^2, \quad U_{\text{cell},2,\text{stat}} = 30.788 \quad (15)$$

is minimized s.t. PDAE/integro-DAE (1–12) and control constraints $u(t) \in U$.

In the discretized version the PDAE/integro-DAE constraint (1–12) is replaced by the semi-explicit DAE (13) and then solved by the software package NUDOC-CCS (Büsken [1]) which transforms this problem into a nonlinear programming problem which is finally solved by a SQP algorithm.

Since this surrogate problem is still too time consuming, a slightly modified problem is solved. A sequence of optimal control problems

$$\left(\min \int_{t_k}^{t_{k+1}} L dt \text{ s.t. (13) and } u(t) \in U \right)_{k=1,\dots,5} \quad (16)$$

is solved. A logarithmic type grid $t_1 = t^* = 0, t_2 = 0.1, t_3 = 1.1, t_4 = 11.1, t_5 = 111.1, t_6 = t_f = 1111.1$ is used, due to the different time scales of the variables. Initial conditions for the first optimal control problem are the stationary solution for $I_{\text{cell},1}$. Initial conditions for the k -th optimal control problem are the free final conditions of the $(k-1)$ -th optimal control problem. In each time interval $[t_k, t_{k+1}]$ an equidistant control grid of 21 points is used. Spatial discretization is 3×3 .

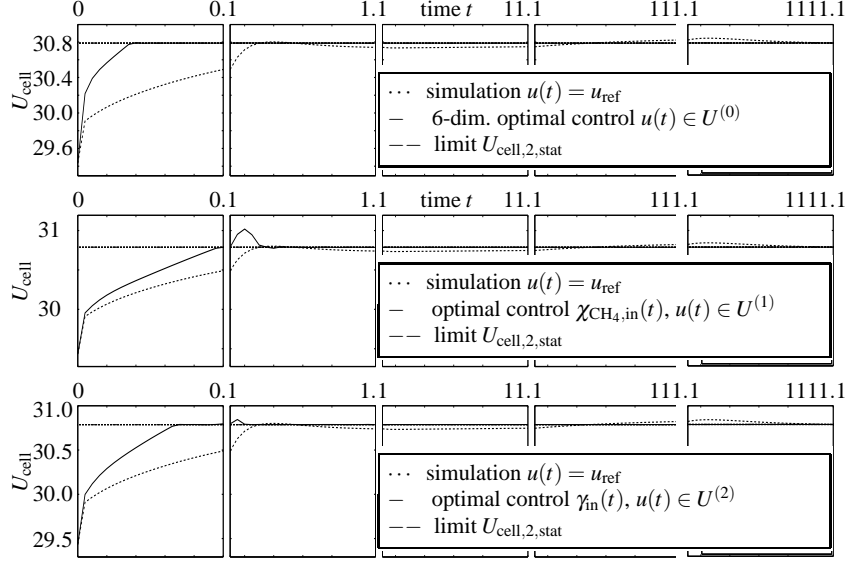


Fig. 3 Simulated and (sub)optimal controlled cell voltage U_{cell}

Numerical solutions are presented for seven different admissible control sets $U^{(0)} \stackrel{\text{def}}{=} \{u \in \mathbb{R}^6 \mid \underline{u}_i \leq u_i \leq \bar{u}_i, i = 1, \dots, 6\}$, $U^{(p)} \stackrel{\text{def}}{=} U^{(0)} \cap \{u \in \mathbb{R}^6 \mid u_i = u_{i,\text{ref}}, i \neq p\}$, $p = 1, \dots, 6$, see Table 1 and Figure 3 and 5. The other molar fractions at the anode inlet fulfill $\chi_{\text{H}_2\text{O},\text{in}} = 1 - \chi_{\text{CH}_4,\text{in}}$, $\chi_{k,\text{in}} = 0$, $k \notin \{\text{CH}_4, \text{H}_2\text{O}\}$. Computational time for the (sub)optimal control with a 6-dimensional control vector $u(t) \in U^{(0)}$ is about 3 days, for the other scenarios with a scalar control about 2 hours. Fig. 3 and 5

Table 1 Control constraints and reference values

	\underline{u}_i	$u_{i,\text{ref}}$	\bar{u}_i
$u_1 = \chi_{\text{CH}_4,\text{in}}$	0.25	1.0/3.5	0.4
$u_2 = \gamma_{\text{in}}$	0.85	1.0	1.5
$u_3 = \vartheta_{\text{in}}$	2.8	3.0	3.2
$u_4 = \lambda_{\text{air}}$	1.5	2.2	2.5
$u_5 = \vartheta_{\text{air}}$	1.0	1.5	2.5
$u_6 = R_{\text{back}}$	0.4	0.5	0.6

present the cell voltage $U_{\text{cell}}(t)$ on the five time intervals $[t_k, t_{k+1}]$.

A fast increase of the cell voltage can be seen in the simulation until $t \approx 0.005$. This is the immediate consequence of the very fast change of the electrical variables. A moderate increase until $t \approx 0.015$ is due to the fast change of the molar quantities. The final stationary value is reached only after about $t \approx 1000$ (over 3 hours),

due to the slow changes in the solid temperature ϑ_s . The oscillating behavior while reaching the new stationary cell voltage has undesirable effects on the cell power $P_{\text{cell}} = I_{\text{cell}}U_{\text{cell}}$ and should be avoided.

The new stationary cell voltage is reached significantly earlier in the optimal control scenarios. For the 6-dimensional control function the cell voltage is in a 0.1%-tube around the new stationary solution after $t \approx 0.03$.

The following scenarios examine, whether the technological expensive control of all input variables is really needed. Especially the scalar control of the molar fraction $\chi_{\text{CH}_4, \text{in}}$ or of the molar flow density γ_{in} give promising results, see Fig. 3. Most favorable seems the scalar control of the molar flow density γ_{in} : the excess cell voltage in the second time interval is lower. Moreover this approach can be easily realized in practice. The scalar optimal control γ_{in} is given in Fig. 4. Fig. 5 presents the less efficient other scalar optimal control scenarios for reference purposes.

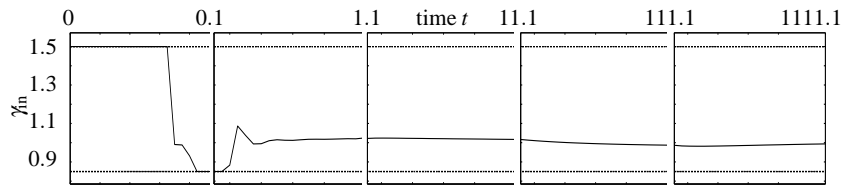


Fig. 4 Scalar (sub)optimal control γ_{in}

4 Conclusion and Outlook

Several suboptimal boundary control strategies were numerically computed and compared for a complicated partial differential algebraic equation system modelling realistically the dynamical behavior of a molten carbonate fuel cell. Although technologically relevant optimal control results could be computed, the huge computational time needed demands model reduction techniques, which are currently under development.

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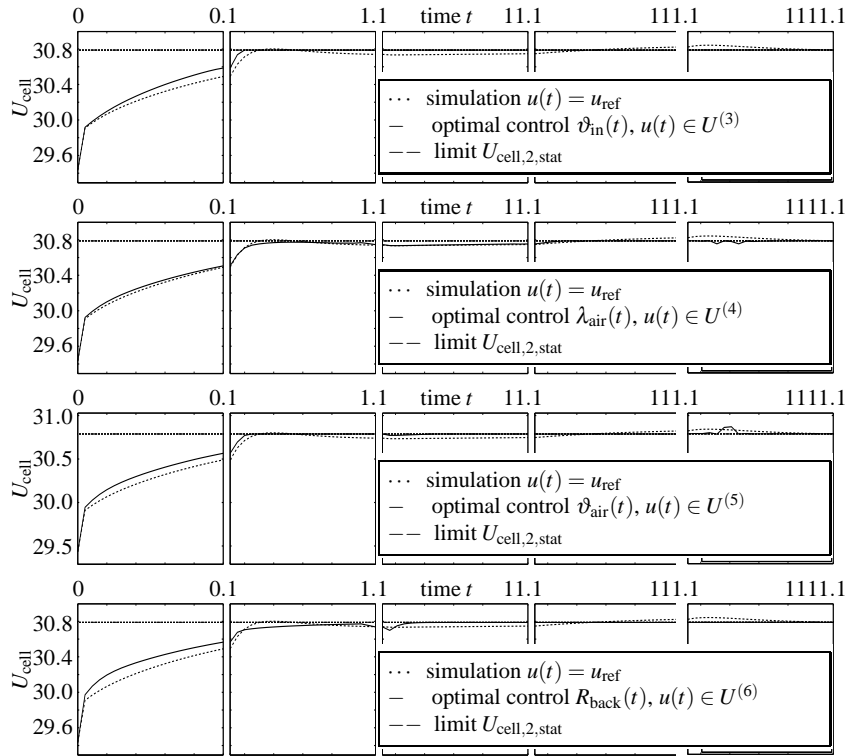


Fig. 5 Simulated and (sub)optimal controlled cell voltage U_{cell}

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