Modeling, Simulation and Optimization of a Cross Flow Molten Carbonate Fuel Cell

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Introduction

The molten carbonate fuel cell (MCFC) consumes fuel gases containing hydrogen, carbon monoxide and light hydro-carbons to produce electric energy. In addition to its high electric efficiency, its operating temperature of about 600°C makes the MCFC a suitable candidate for the coupled production of electricity and heat in stationary applications. Due to its insensitivity with respect to carbon monoxide, it is very flexible with respect to fuels making the MCFC suitable for a broad range of applications.

In Germany, the company MTU CFC Solutions has developed a 250 kW$_e$ MCFC system, called the "HotModule" (Figure 1, [1]). Its electric system efficiency of about 50% is unsurpassed by conventional systems in this power range, and it has proven its feasibility and reliability in more than 20 successful field trial plants in Germany, Europe, Japan and the US. Currently, the project is heading towards commercialization, with the planned start of a series production of economically competitive systems in 2006.

Although the HotModule has proven its reliability, there are still some optimization potentials left. Currently the HotModule applies external, indirect internal and direct internal reforming, leading to a high hardware effort. Furthermore, the design of these three reforming stages is not balanced against each other. A detailed design optimization can help to reduce building costs while increasing the system performance.
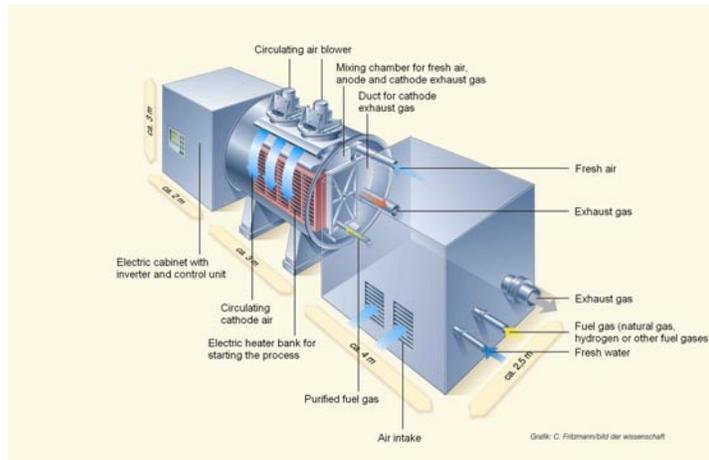
Figure 1: Schematic of the HotModule by MTU CFC Solutions, Germany ([1,2]).

Another aspect is the temperature field in the cell stack, which is of crucial importance to the life time and performance of the cell. Material properties limit the maximum temperature; thermal stress and thermal expansion limit the maximum allowable temperature difference in the stack. Measurement informations on this very important state are incomplete. As a consequence, operating conditions are fixed based on empirical

values and contain large safety factors, leading to sub-optimal efficiency. Numerical optimization of operating conditions with respect to the given constraints can significantly contribute to the safe and efficient operation of the cell.

Besides optimization, system control is of vital interest for the operation of a real HotModule. An online state observer can monitor important system states which are not directly measurable and can yield information for a control system for disturbance rejection. With respect to dynamic load changes, except for emergency shut-downs, the electric load at the HotModule is altered only in small steps, so large power demand increase can not be realized quickly. Thus, another promising aim is to find safe and optimal load change strategies and tracking controls.

These issues are the objectives of a joint research project of three academic and two industrial partners funded by the German Ministry of Education and Research. These are the University Magdeburg, the Max-Planck-Institute Magdeburg and the University Bayreuth together with the MTU CFC Solutions and the IPF Heizkraftwerkbetriebsgesellschaft, which is operating a HotModule at the university hospital in Magdeburg.

All of these aims require mathematical cell models of differing detail level and complexity. To reduce the modeling effort, one single reference model is derived, from which all other required models are obtained either by physical simplifications or by mathematical reduction methods (see Figure 2). The advantages of this hierarchical modeling strategy are clear: the overall modeling effort is reduced and the individual model variants are comparable to each other. Results obtained from one model can be transferred to another one more easily, and system parameters in different models have identical physical meanings.

In the following, we give a look into the properties and purposes of each of the models. Additionally, we show some examples of how these models are used to solve the mentioned issues concerning MCFC.

The reference model

The reference model basically describes a spatially two-dimensional single MCFC in cross-flow configuration at galvanostatic operating mode (see Figure 3). In addition to this cell, a catalytic combustion chamber is considered. The model is based on the balances of mass, charges and energy and therefore fulfills the corresponding laws of conservation. It allows the simulation of the compositions and flow density inside the anode and cathode gas channels, the temperature distributions in both gas phases and in the solid parts of the cell, and also the most important electric quantities, i.e. the cell voltage and current density distribution. It is completely formulated in terms of dimensionless parameter groups and variables [3,4].

The equation system contains a number of hyperbolic partial differential equations (PDE, describing the concentrations and temperatures in the gas phases), one parabolic PDE (describing the temperature in the solid cell parts), several ordinary differential equations (ODE), and one integral equation (defining the total cell current as the integral of the cell current density). Additionally, there are a few implicit algebraic equations (AE, one of them is the enthalpy balance of the combustion chamber) and a large number of explicit AE.

Especially due to the implicit AE, the numerical treatment of this model is rather slow, making it unsuitable for most applications. Thus, the sole purpose of this model is to serve as a reference basis for the derivation of further models, which are adapted to the various aims in our project.
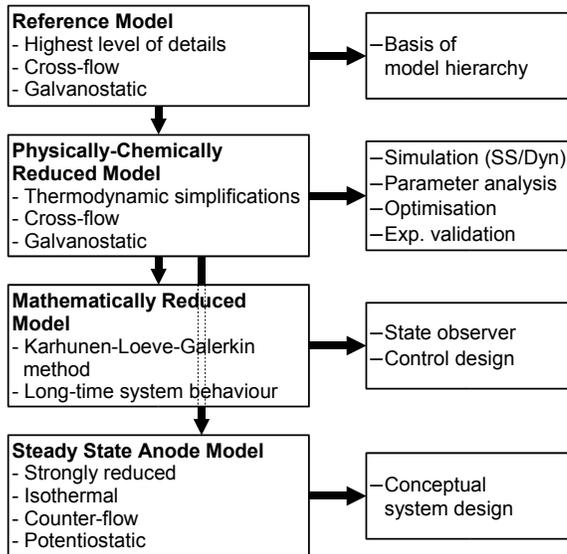
Figure 2: Model hierarchy.
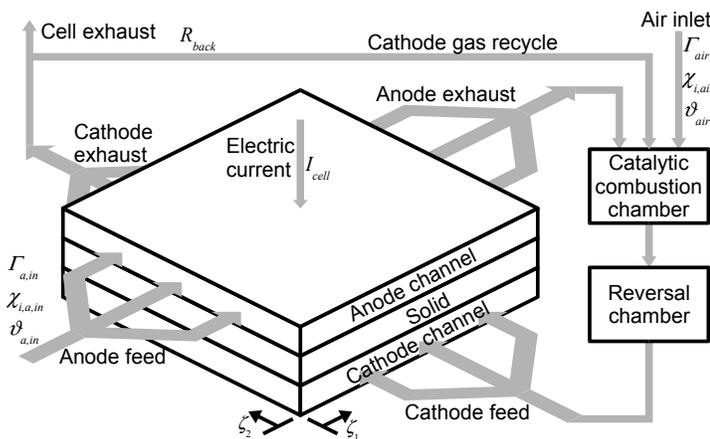
Figure 3: Compartments and fluxes of the reference model consisting of anode and cathode gas layer, a solid phase, a catalytic combustion chamber and a reversal chamber. Also indicated are the input parameters of the system.

Physically-chemically reduced model

The physically-chemically reduced model is derived by applying a few minor thermodynamic simplifications to the reference model. This model has a similar structure as the reference model, but the simplifications allow for a significantly faster numerical solution. Within our project, this model together with its variants are most frequently used.

As an example for a steady state result, Figure 4 shows the spatial temperature distribution inside the solid parts of the cell under certain load conditions. This distribution is typical for high temperature fuel cells, with the maximum temperature at the outlet/outlet corner of the anode and cathode gas channels and minimum temperature in the opposite corner. The reason for this is the convective transport of heat by both gas phases, which

are heated up by the solid cell parts along the channels. It is this temperature which may not exceed certain values in order to avoid mechanical damage to the cell stack.
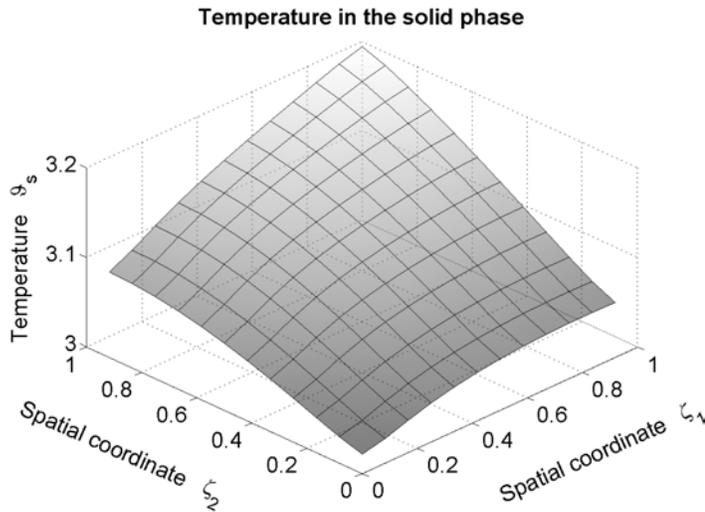
Figure 4: Steady state temperature profile inside the cell's solid parts. The temperature range displayed here corresponds to a range from 894 K (621°C) to 954 K (681°C). Note that the anode gas flows along the ζ_1-coordinate, while the cathode gas flows along ζ_2.

When applying a load change to the system, the cell voltage is of high interest. Figures 5 and 6 show the short-term and the long-term behavior of the cell voltage after a stepwise increase of the electric load which is demanded from the cell. In this simulation, only the electric load is increased, but all other input parameters like amount and composition of the feed gas are kept constant. At the time of the load change, the cell voltage drops down instantaneously. During the next few seconds, it decreases even further due to the depletion of educts in the cell channels (Figure 5). This is called the short-term behavior. After a few seconds, the concentrations reach a new quasi steady state and the dynamics of all processes in the cell are enslaved by the very slow dynamics of the cell
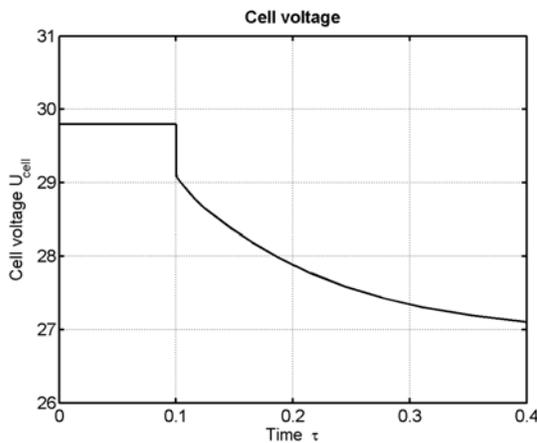
Figure 5: Short-term behavior of the cell voltage over the first few seconds after a stepwise load change at τ=0.1. A dimensionless cell voltage of U_{cell}=38.9 corresponds to 1 Volt.
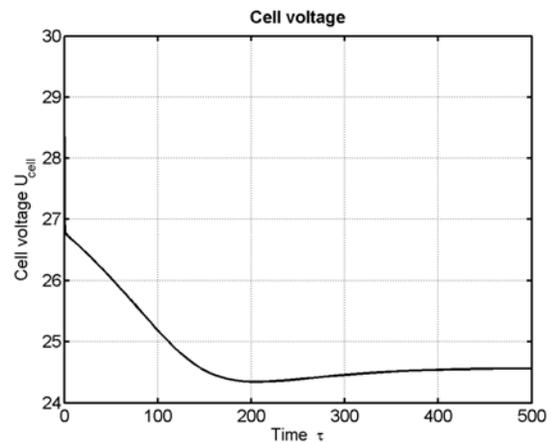
Figure 6: Long-term behavior of the cell voltage over several hours after a stepwise load change. A dimensionless cell voltage of U_{cell}=38.9 corresponds to 1 Volt.

temperature change. This phenomenon can be seen in Figure 6, where the cell voltage changes over several hours due to slow temperature change. In the case shown here, the cell voltage even runs through a temporary minimum before finally reaching its new steady state. The discrimination between short-term and long-term behavior is essential for the control design.

Besides the prediction of states inside the cell the physically-chemically reduced model can also be applied for optimization. Here, two different studies have been performed. First, optimal operating conditions are evaluated for different electric loads. The result are points of an optimal voltage-current curve, which represent the operatings conditions with the highest possible efficiency at given load. Figure 7 shows the corresponding electric system efficiency over the cell current. In addition, the optimization considers several temperature constraints, so that the operating conditions also guarantee safe operation.

The second optimization also includes system design. The idea is to utilize the spatial distribution of the reforming catalyst density inside the anode channels in order to homogenize the temperatures and increase the system efficiency. As a result, we obtain an optimal reforming catalyst distribution, which allows us to operate the cell at higher efficiencies than with homogeneous catalyst loading along the anode channels (see Figure 7).
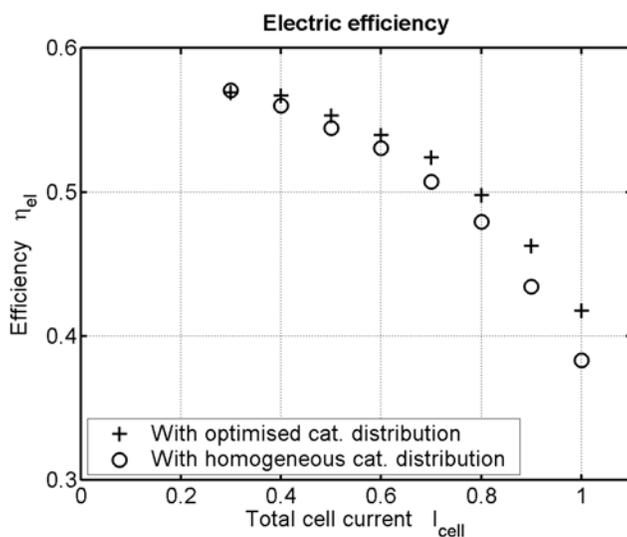
Figure 7: Electric efficiencies for a system with homogeneous reforming catalyst density and with optimal catalyst density distribution as a result of the optimizations of operating conditions and design parameters.

Mathematical model reduction and application to a model based measuring system

The numerical solution of the physical model introduced in the last section requires a large amount of computation time. Therefore, this model is less suitable for process control purposes and real-time applications. A reduced model formulation is desirable that approximates the original model with reasonable accuracy but consists of an equation system of low order and is more easily accessible to numerical and analytical treatment. Such a reduced model was derived in a two-step procedure: In the first step, the original model was simplified by assuming quasi-steady state for mass and charge balances. In the second step, the Karhunen-Loève Galerkin method was applied to this simplified model.

Details of this model reduction process are found in [5]. The resulting reduced model consists of a system of ordinary differential equations and algebraic equations. As can be seen from Figure 8, the reduced and the original model agree well already for a small number of basis functions used in the Galerkin approach. In terms of system order, a model reduction by a factor of 1000 can be achieved. The reduction in terms of CPU time is by a factor of about 100 and not quite as strong as the reduction of the system order, because the evaluation of the reduced model equations requires a numerical quadrature and therefore is more complicated.
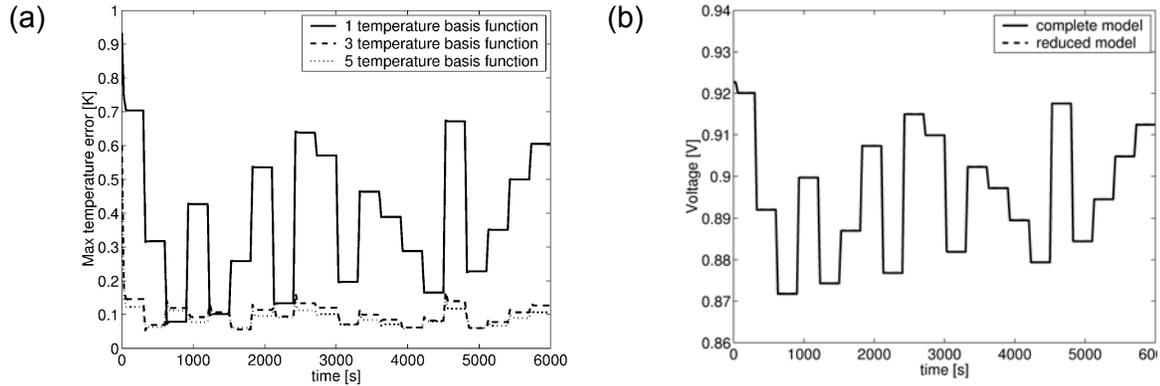
Figure 8: Validation of the reduced model by a test simulation with randomly varying cell current; (a) maximum temperature error of the reduced model; (b) cell voltage of reduced and original model.

The reduced model can be applied in the framework of a model based measuring system [6]. A model based measuring system or state observer estimates states not directly measurable by doing an on-line simulation parallel to the real process. It consists of a simulator part – the reduced fuel cell model – and a corrector part that compares simulated and real measuring values and compensates for model errors and unknown disturbances (see Figure 9). Uncertain model parameters, e.g. kinetic coefficients, can be included in the estimation process by defining dummy equations for them.
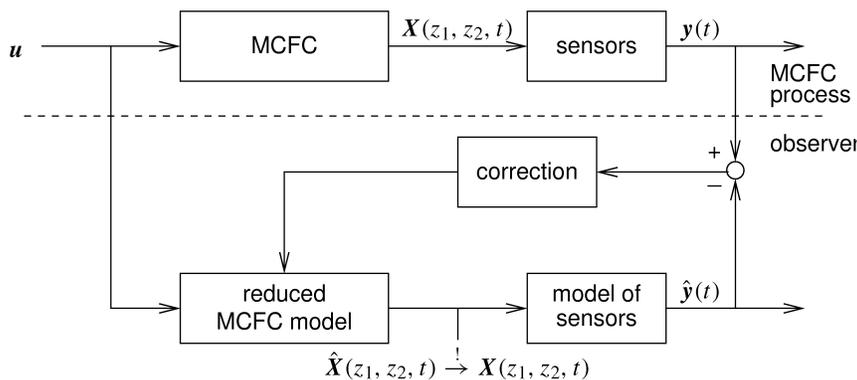
Figure 9: Block diagram of the model based measuring system for the MCFC.

A test simulation of the state observer is shown in Figure 10. Initially, the observer model contains a wrong value for the pre-exponential factor of the anodic reaction k_0^A. Consequently, the steady state of the reduced model deviates strongly from the correct

solution. After some time, however, the observer corrects the wrong initial conditions and the wrong parameter value and gives a good estimate of the real process.
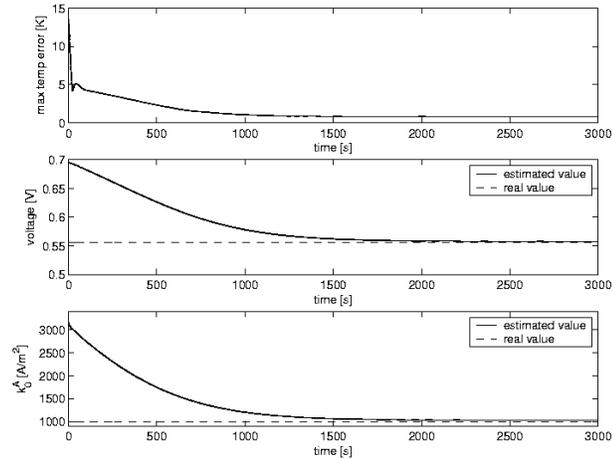
Figure 10: Simulation results of the model based measuring system based on the reduced model (simulation results from the physical model used as measuring values). Top diagram: maximum temperature difference between observer and real system; middle diagram: estimated and real cell voltage; lower diagram: estimated and real kinetic parameter.

Steady state anode model

From the physically-chemically reduced model, another model can be derived by applying several simplifications concerning the enthalpy balance, reflux ratio and the reaction system involved in the system. This yields the steady state anode model which describes the gas composition in the anode channel in terms of extents of reaction by two ODE [7]. The simplicity of the models allows for fast numerical solution whose results can be illustrated in a compact conversion diagram as given in Figure 11.

The applications of this model range from the comparison of different reforming concepts like external, indirect and direct internal reforming, to the design of fuel cell cascades, the benefit of which can be estimated quantitatively by the steady state anode model. Figure 11 shows the conversion diagram for a single cell together with that of a two-cell cascade of equal size. The positive horizontal axis shows how far the reforming process proceeded, while the extent of the oxidation reaction can be seen at the vertical axis, which is a measure for the electric current produced. The applied cell voltage is plotted along the left hand side of the horizontal axis. The area of the rectangle created by the extent of oxidation reaction and the cell voltage is a measure for the electric cell power produced by the cell. By optimization of the cell voltages, the performance of the two systems is maximized. It turns out that the two-cell cascade offers a higher electric power output compared to the single cell configuration.
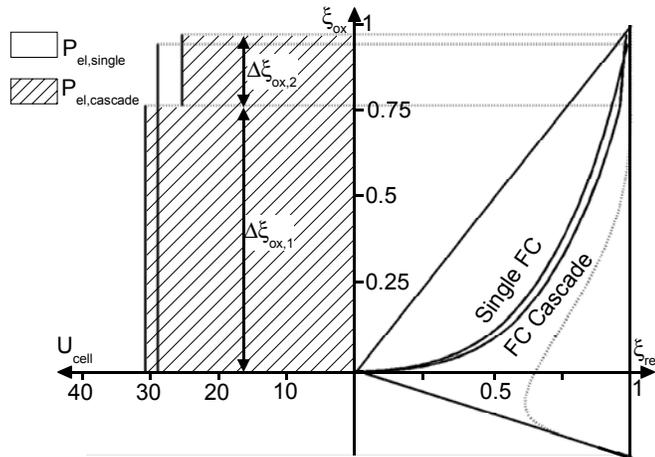
Figure 11: Conversion diagram for a single cell and a two-cell cascade of equal size. The overall electric power output of the cascade is higher, as indicated by the area of the rectangles on the left hand side.

Conclusions

This contribution gives an overview on ongoing research activities on the simulation, optimization and control design of molten carbonate fuel cells (MCFC). Motivated by several remaining issues related to design and operation of MCFCs we set up a hierarchy of models. Four of them are presented here and their contributions to threat different aspects concerning the MCFC are demonstrated, ranging from conceptual design over optimizations to observer and control design.

Acknowledgements

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