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Multiscale CFD simulation of a methane steam reformer for optimization of the spatial catalyst distribution

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Abstract

This paper studies the coupled mass and heat transport as well as the reactions in an Indirect Internal Reforming (IIR) unit of a Molten Carbonate Fuel Cell (MCFC). The aims are first to identify the dominating processes for a specific design. Because temperature is one major issue in MCFC, the second aim is to predict the spatially distributed thermal behavior of the unit. In a first step several detailed models, describing only a small section of the IIR, are created. The results of these simulations, especially the temperature and gas composition, are discussed. In a second step the whole IIR unit is modeled. The results of the detailed models and some simplifications of the geometry are used to create this model.

Keywords

reactor engineering, multiscale simulation, Molten Carbonate Fuel Cell (MCFC), Internal Indirect Reforming (IIR), CFD

1. Introduction

The fuel cell technology is based on the work done by Christian Friedrich Schönbein and Sir William Grove 150 years ago. Since that time different types of fuel cells were and are developed. One of them is the Molten Carbonate Fuel Cell (MCFC).

In a MCFC methane is used as fuel. It has to be reformed, which can be done in the Indirect Internal Reformer (IIR). The IIR is a flat reactor located between the single cells of the fuel cell stack. Within this reactor the methane reforming as well as the water gas shift reaction take place.

methane reforming: $CH_4 + H_2O <=> CO + 3\ H_2$
water gas shift reaction: $CO + H_2O <=> CO_2 + H_2$

The reaction products CO and H_2 are used as fuel in the electrochemical reactions. Further on the temperature distribution of the fuel cell stack is influenced by the endothermic reactions within the IIR. The temperature in turn is one of the important values to describe the state of the fuel cell stack. The reaction rates (efficiency) as well as the material degradation (life time) depend on the temperature [1]. Therefore a model of the IIR is an important tool for the optimization of the temperature distribution within the fuel cell stack.

2. Multiscale modeling

The structure of the IIR unit is shown in Fig. 1 [2]. The flow field within the reactor is enforced by a corrugated sheet. The repeating pattern of the sheet creates rows in which the cylindric catalyst pellets can be filled (see the zoom view in Fig. 1). The IIR unit is divided in four sections with different flow directions and different spatial catalyst distributions:

I.   On the right hand side the gas goes bottom-up in the IIR unit. There is no catalyst within this section.

II.   Following the orientation of the corrugated sheets, the gas flows in this section from the right to the left. Here 1 of 4 rows is filled with catalyst pellets.

III. Also in the third section 1 out of 4 rows is allocated with catalyst pellets. In this section the flow direction is top down.

IV.   In the last section the flow direction stays top down but the catalyst allocation is changed to 2 out of 4 rows.

The modeling of the IIR unit is done in two steps. In a first step, detail models of a small section are created taking into account the exact 3D geometry. In these models catalyst allocations of 1, 2 and 3 out of 4 rows are analysed. The knowledge from these detailed simulations is used to create a model of the entire IIR unit [3]. All simulations are done using the CFD tool CFX 10.0.
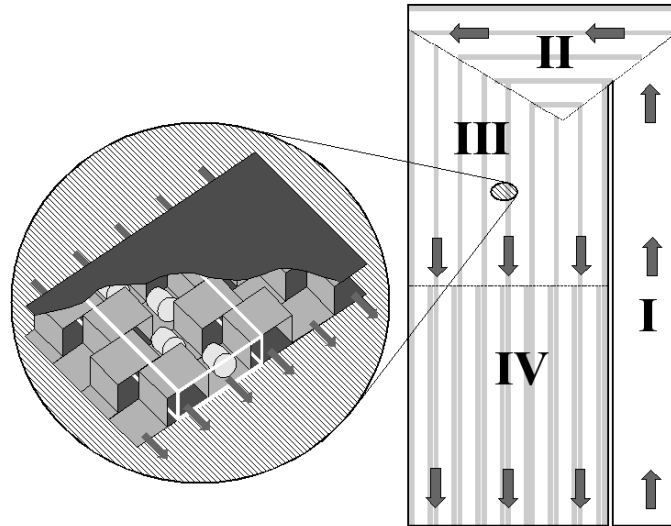
Fig. 1: Schemata of the entire IIR unit (US-Patent 6200696) with gas flow directions and a detailed view of the structure. In the zoom view the corrugated sheet is shown. One row filled with catalyst pellets is indicated by the white frame.

3. Detail models

For the detail models a small part of 4 rows width and 15 pellets length is cut out from the IIR unit. An inlet and an outlet section are added. The known balance equations for incompressible flow are used [4]:

- the incompressible Navier-Stokes equation with a k-ε turbulence model,
- the energy balance equation,
- the mass balance and
- the component balances for CH_4, CO, CO_2, H_2O and H_2.

For the corrugated metal sheet as well as for the top and bottom sheet the heat transfer equation is taken into account.

At the inlet boundary all properties of the gas are set whereas at the outlet boundary the reference pressure is given. For the top and bottom sheet a constant temperature is assumed. The right and left border of the model are connected using a periodic boundary condition.

For both reactions a simple power law is used including the thermodynamical equilibrium. The reactions take place only at the surface of the catalyst pellets.

$$r_{ref} = k_{0,ref} \left[\left(\frac{p}{p^0} \right)^2 x_{CH_4} x_{H_2O} - \left(\frac{p}{p^0} \right)^4 \frac{1}{K_{ref}(T)} x_{CO} x_{H_2}^3 \right] \qquad (1)$$

$$r_{wgsr} = k_{0,wgsr} \left(\frac{p}{p^0} \right)^2 \left[x_{CO} x_{H_2O} - \frac{1}{K_{wgsr}(T)} x_{CO_2} x_{H_2} \right] \qquad (2)$$

3.1. Results for a catalyst allocation of 1 out of 4 rows

Fig. 2 shows the methane concentration, the temperature distribution and the velocity for a detail model with 1 out of 4 rows filled with catalyst pellets. Due to the reactions taking place at the surface of the catalyst pellets a temperature drop can be observed close to the pellets. One can clearly distinguish between a reactive zone where the temperature changes and a non-reactive zone where the temperature stays at a constant value. Within the simulated section of the IIR unit a constant pattern for the temperature distribution is reached. This indicates that the heat needed for the reaction is balanced by the heat flow through the top and bottom sheet.

For the methane molar fraction a similar distribution can be seen. At the surface of the catalyst pellets methane is consumed by the reforming reaction. Within the first few pellets the chemical equilibrium is reached around the pellets. Due to turbulence and diffusion the reaction products are transported away from the pellet surface and the educts are transported towards the reactive zone. This indicates that after a few pellets the reactions are independent from the catalyst activity and are only limited by the mass transfer.
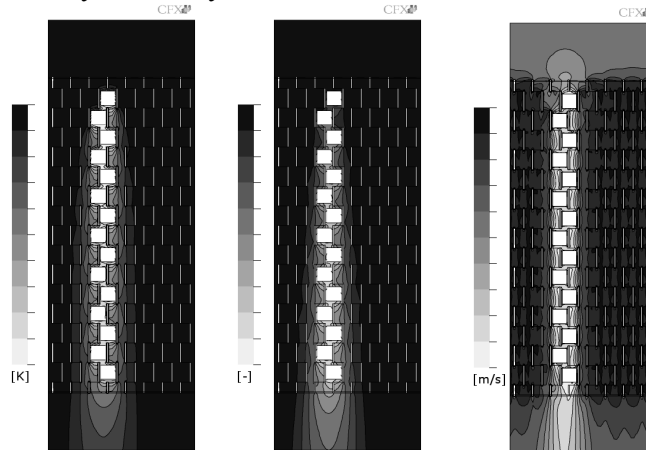
Fig. 2: Detail model with 1 out of 4 rows filled with catalyst pellets. Left: temperature; Middle: methane molar fraction; Right: velocity; Black indicates higher values whereas white indicates lower values.

3.2. Comparison of the results for different catalyst allocations

Fig. 3 shows the methane molar fraction for an allocation of 1, 2 and 3 out of 4 rows filled with catalyst pellets. The reactive and non-reactive zones are clearly distinguishable. Next to the catalyst pellets the chemical equilibrium is reached and there is a mass transfer limitation for the reactions.
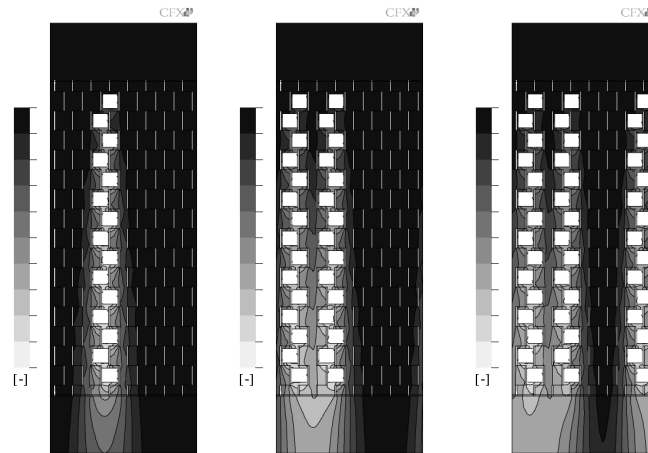

Fig. 3: Methane molar fraction within the detail models for 1, 2 and 3 rows filled with catalyst pellets. Black indicates higher methane molar fraction whereas white indicates a lower methane molar fraction.

4. Model of the entire IIR reactor

For the simulation of the entire IIR unit the geometry is simplified. A quasi-homogenous model is used where the pressure drop due to the pellets and the corrugated sheet is simulated using an anisotropic permeability. A locally distributed catalyst activity incorporates the results of the detail models.

Fig. 4 shows the methane molar fraction and the temperature distribution within the entire IIR unit. There are no catalyst pellets in the first part of the model (section I) so the methane molar fraction does not change. At the entrance to section II one can observe a drop in the methane molar fraction. The pattern of the rows filled with catalyst pellets is clearly visible in section II to IV.

In the plot of the temperature distribution this pattern can also be found. Where there are catalyst pellets the endothermic reaction cools the gas. This cooling effect is especially visible at the border between section I and section II due to the high methane molar fraction and the high temperature. At the border between section II and section III one can also observe a lower temperature, but here it is due to the change in the flow direction.
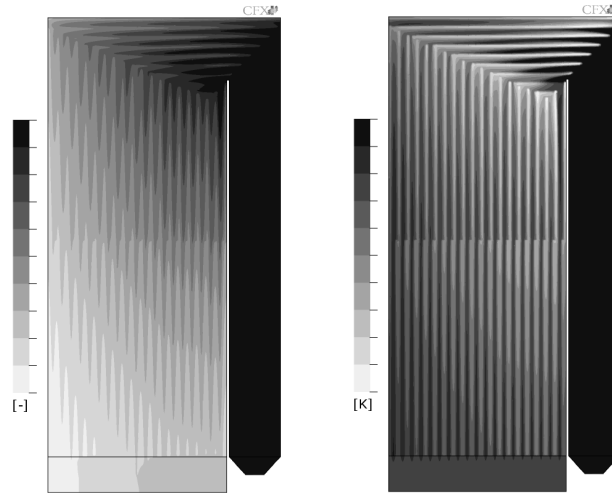
Fig. 4: Methane molar fraction (left) and temperature distribution (right) within the model of the entire IIR unit. Black indicates higher values whereas white indicates a lower methane molar fraction or temperature, respectively.

5. Conclusions

The simulation of the IIR unit was done in two detail levels. With the detail level the basic effects of the 3D geometry of the corrugated sheet and the catalyst pellets could be analysed. The reactions do not depend on the catalyst activity but are only limited by mass transfer. Connecting this IIR model with a model for the neighboring fuel cells could make optimization of the temperature profile possible by optimization of the catalyst distribution within the IIR unit.

References

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