Thermal Modeling of a Honeycomb Reformer including Radiative Heat Transfer

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Abstract: Reformer and catalytic burners are common components in fuel cell systems, crucial for efficient preparation of fuel and exhaust gases of the fuel cell stack. In this abstract we intend to show the influence of radiation to the temperature distribution inside of a cylindrical reformer unit. The model consists of an axisymmetric representation of the inlet-zone and a catalytic porous zone. Fluid flow, convective and conductive heat transfer and simplified chemical reactions for oxidation and reforming are included as physical models. Due to temperatures up to 900°C in the reactor radiation should be considered. For the inlet zone the Surface to Surface radiation model of COMSOL is used and is coupled to a 1D user defined radiation in participating media model for the honeycomb which consists of various small channels. Temperature distributions for models with and without radiation are compared.

Keywords: radiation model coupling, reforming, reactor model, sofc

1. Introduction

Catalytic reforming is a common process in fuel cell systems to produce synthesis gas from methane. Common partial oxidation reforming reactors consist of a premixing zone where methane and air are prepared, a chamber for further mixing and the broadening of the fluid flow for entering the catalytic honeycomb channel structure. Inside of the honeycomb, oxidation and reforming reactions take place, generating hydrogen from methane and oxygen which leaves the reforming unit and enters the fuel cell stack.
The oxidation of methane leads to local high temperatures mainly responsible for catalyst aging. Selective experimental measured temperatures are ranging from 300°C up to 900°C and radiation therefore should be included. Beside of the catalyst aging also the chemical equilibrium of species concentrations depends on local temperatures hence the focus of this model is on the temperature distribution.

2. Set Up and Governing Equations

Figure 1 shows the setup up of the considered reformer geometry with the inlet pipe of 6mm cross-section, next to the mixing geometry (not shown here), ending in a pipe with 31mm diameter where the honeycomb is located 30mm behind the cross-section is rising. The catalyst particles are located on a 400cpsi cordierite support and the cross section of one channel is 1.1x1.1mm^2.

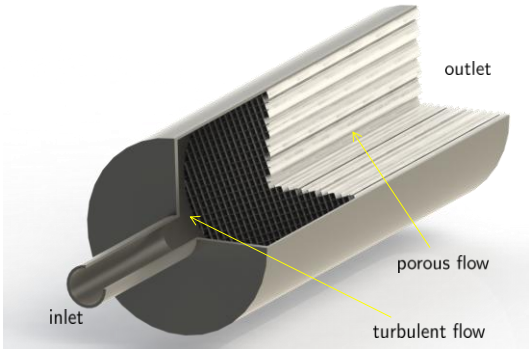

Figure 1. Setup up of the reformer unit.

2.1 Fluid Flow

There are two different flow regimes denoted in Figure 1, a free-flow inlet region, where an abrupt rise of the cross-section occurs and a porous flow region inside of the honeycomb. For the free-flow region the well known k-ε turbulence model (1) is applied.

$$\rho(\vec{u}\cdot\nabla)\vec{u} = \nabla\cdot\left[-p\hat{I} + (\mu+\mu_t)\left(\nabla\vec{u} + (\nabla\vec{u})^T\right) - \frac{2}{3}(\mu+\mu_t)(\nabla\cdot\vec{u})\hat{I} - \frac{2}{3}\rho k\hat{I}\right] \quad (1)$$

$$\nabla \cdot (\rho \vec{u}) = 0 \tag{2}$$

$$\rho(\vec{u} \cdot \nabla)k = \nabla \cdot \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \nabla k \right] + P_k - \rho\varepsilon \tag{3}$$

$$\rho(\vec{u} \cdot \nabla)\varepsilon = \nabla \cdot \left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \nabla \varepsilon \right] + C_{\varepsilon 1} \frac{\varepsilon}{k} P_k - C_{\varepsilon 2} \rho \frac{\varepsilon^2}{k} \tag{4}$$

$$P_k = \mu_t \left[\nabla \vec{u} : \left(\nabla \vec{u} + (\nabla \vec{u})^T \right) - \frac{2}{3} (\nabla \cdot \vec{u})^2 \right] - \frac{2}{3} \rho k \nabla \cdot \vec{u} \tag{5}$$

In the above equations ρ, μ, μ_t, p, P_k k, ε and $\vec{u}$ are the density, the kinematic viscosity, turbulent viscosity, pressure, corrected pressure, turbulent kinetic energy, turbulent dissipation rate and the local velocity. The density is obtained by the ideal gas law

$$\rho = M \cdot \frac{p}{R T_{gas}} \tag{6}$$

where M, R and T_{gas} are the mean molar mass, universal gas constant and the gas temperature. The turbulent viscosity is calculated from

$$\mu_t = \rho C_\mu \frac{k^2}{\varepsilon} \quad . \tag{7}$$

The constants in equation 1 $C_{\varepsilon 1}, C_{\varepsilon 2}, C_\mu, \sigma_\varepsilon$ and σ_k are the common parameters for the k-ε turbulence model.

The catalytic honeycomb consists of many small channels and for each a laminar flow can be assumed where the pressure drop can be calculated by Darcys law.

$$\vec{u} = -\frac{\hat{\kappa}}{\mu} \cdot \nabla p \quad . \tag{8}$$

Using an anisotropic permeability $\hat{\kappa}$ for the whole honeycomb makes it possible to use an axis-symmetric geometry for the reformer unit model and to lower computational costs.

2.2 Pseudo Reactions

For testing purposes two pseudo reactions are included which should reproduce a fast exothermic and respectively a slower endothermic reaction heat release. This is achieved by using the following equations:

$$\begin{aligned} \nabla(\rho \vec{u} \phi_{oxi}) &= -\rho k_{oxi} \phi_{oxi} \\ \nabla(\rho \vec{u} \phi_{ref}) &= -\rho k_{ref} \phi_{ref} \end{aligned} \tag{9}$$

where k_{oxi} and k_{ref} are the rate constants of either the oxidation or the reforming reaction.

The variables ϕ_{oxi} and ϕ_{ref} are fixed to a value of one at the reformer inlet and the right hand side of (9) is only different from zero inside of the honeycomb. The corresponding heating rate is defined by:

$$\dot{q}_{reac} = \left(P_{oxi} k_{oxi} \phi_{oxi} + P_{ref} k_{ref} \phi_{ref} \right) \frac{\rho}{\dot{m}} \tag{10}$$

where P_{oxi} and P_{ref} are the total exothermic reaction power and the total needed endothermic reaction power, which are chosen so that the output temperature reaches 850°C. The quantity $\dot{m}$ denotes the total mass flow rate.

2.3 Heat Transport

2.3.1 Convection and Conduction

Inside of the reforming unit the convection and conduction of heat is treated by:

$$\begin{aligned} \nabla \cdot \left(-\hat{\lambda}_{gas} \nabla T_{gas} \right) + C_p \rho \vec{u} \cdot \nabla T_{gas} = \\ = \alpha_{sol \Leftrightarrow gas} \cdot \left(T_{sol} - T_{gas} \right) \quad . \end{aligned} \tag{11}$$

This equation reproduces the heat transfer in the fuel gas where $\hat{\lambda}_{gas}$, C_p, $\alpha_{sol \Leftrightarrow gas}$ and T_{gas} are the anisotropic heat conductivity of the gas, the mean heat capacity, the volumetric heat transfer coefficient and the temperature of the fuel gas. An additional conductive heat transfer (12) is introduced inside of the honeycomb.

$$\begin{aligned} \nabla \cdot \left(-\hat{\lambda}_{sol} \nabla T_{sol} \right) = \\ = -\alpha_{sol \Leftrightarrow gas} \cdot \left(T_{sol} - T_{gas} \right) + \dot{q}_{reac} + \dot{q}_{rad} \end{aligned} \tag{12}$$

Here $\hat{\lambda}_{sol}$, T_{sol}, $\dot{q}_{reac}$ and $\dot{q}_{rad}$ are the anisotropic heat transfer coefficient of the honeycomb, the solid temperature, the reaction heat source due to oxidation and reforming processes and the radiative heat source

$$\dot{q}_{rad} = \varepsilon_{por} \kappa_{rad} (-2\sigma_{rad} T_{sol}^4 + I_1 + I_2) \tag{13}$$

appearing from absorption and emission of radiation from the channel walls of the honeycomb (see next section). It is assumed that the species concentrations at catalytic surface are equal to gas phase concentrations.

2.3.2 Radiative Heat Transfer

Due to the high temperatures one should account for radiative heat transfer. Between the walls of the free-flow region and the honeycomb entrance, surface-to-surface radiation can be assumed because optical thickness is thin in respect to distances inside of the reforming unit. Therefore one can use the COMSOL module Surface-to-Surface Radiation as it is. Inside the honeycomb a participating media approach for the homogenized channel structure is needed but the build-in COMSOL module includes only isotropic absorption. So an anisotropic 1d participating media approach is introduced by including the following PDEs:

$$+\frac{dI_1}{dz} = \kappa_{rad}\sigma_{rad}T_{sol}^4 - \kappa_{rad}I_1 \qquad (14)$$

$$-\frac{dI_2}{dz} = \kappa_{rad}\sigma_{rad}T_{sol}^4 - \kappa_{rad}I_2 \qquad (15)$$

Intensity I_1 is starting from the honeycomb inlet and intensity I_2 is starting from the outlet of the honeycomb. Both intensities are coupled via the local solid temperature. The absorption coefficient κ_{rad} was determined from a separate 3d channel model and the constant σ_{rad} denote the Stefan-Boltzmann constant.

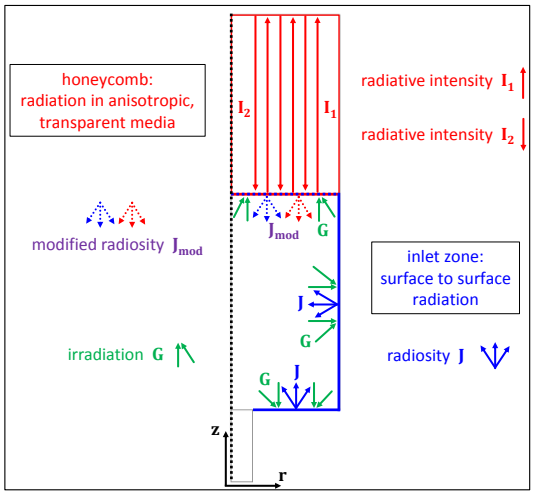

Figure 2. Domains and boundaries with surface-to-surface radiation and radiation in participating media approach.

3. Use of COMSOL Multiphysics

All equations are realized within COMSOL Multiphysics 4.3a.
To solve equations (1) to (5) the Fluid Flow → Single Phase Flow → Turbulent Flow k-ε COMSOL module is used. All boundaries are set to no-slip boundary condition except inlet where mass-flow inlet is chosen and outlet where pressure is set to zero. Within the honeycomb the pressure drop is represented by an additional volume force source term in form of Darcys law (8) so the channel structure is treated as porous media. The appearing anisotropic permeability is determined by analyzing a 2d cross section of one channel but is not shown here. The components of $\hat{\kappa}$ perpendicular to the channel direction are set one order of magnitude lower than in channel direction because they can not be set to zero for numerical reasons.

For the pseudo reaction equation (9) the Mathematics → PDE Interfaces → Coefficient Form PDE COMSOL module is used where the source term f is set to zero except inside the honeycomb.

The heat transfer is introduced by either using the Heat Transfer → Heat Transfer in Fluids COMSOL module for gas temperature and the Heat Transfer → Heat Transfer in Solids COMSOL module for the honeycomb. These modules are coupled by a heat exchange source term $\alpha_{sol\Leftrightarrow gas}\cdot\left(T_{sol}-T_{gas}\right)$. For all presented results adiabatic behavior is assumed thus all boundaries are insulated and only gas inlet is set to a fixed temperature and the outlet is set to convective outflow condition.

For the radiative heat transfer in the free-flow region the Heat Transfer → Radiation → Surface-to-Surface Radiation COMSOL module and inside the honeycomb the Mathematics → Classical PDEs → Convection-Diffusion COMSOL module is used. In Figure 2 all surfaces that are in surface-to-surface radiation exchange are shown. At the honeycomb entrance the surface-to-surface radiation model is coupled to the 1d radiation in participating media and the Heat Transfer in Solids model. This is achieved by introducing a "Prescribed Radiosity

boundary" condition in the Surface-to-Surface radiation mode and the value for radiosity is calculated by:

$$J = \varepsilon_{por} I_2 + (1-\varepsilon_{por})\left[(1-\varepsilon_{rad})G + \varepsilon_{rad}\sigma_{rad}T_{sol}^4\right]. \quad (16)$$

The intensity I_1 is set equal to the irradiation G obtained from the surface-to-surface radiation module and the thermal boundary condition for the honeycomb entrance is given by:

$$-\vec{n}\cdot\left(-\hat{\lambda}_{sol}\nabla T_{sol}\right) = (1-\varepsilon_{por})\varepsilon_{rad}\left(G-\sigma_{rad}T_{sol}^4\right) \quad . \quad (17)$$

At the reformer outlet the intensity I_2 is set to $\sigma_{rad}T_{sol}^4$.

5. Results and Discussion

5.1 Estimating the adsorption coefficient

Comparing the temperature of a detailed 3d model, representing one single honeycomb channel including heat transfer by conduction and surface-to-surface radiation, with a 1d model, ruled by equations (12), (14) and (15) where the right hand side of equation (12) only contains the radiation source term, leads to an estimation for the absorption coefficient κ_{rad}. Figure 3 shows the configuration of the 3d and 1d model with boundary conditions.

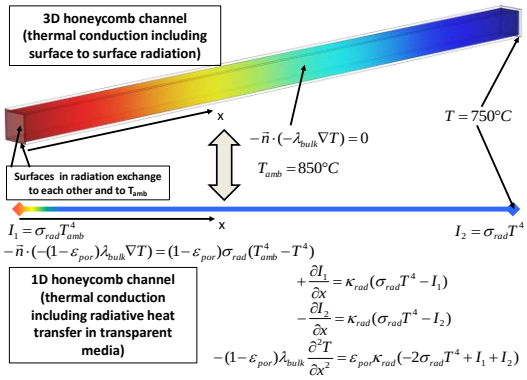

Figure 3. Configuration and boundary conditions for a detailed 3d heat convection model with surface-to-surface radiation and the corresponding homogenized 1d heat conduction model including a radiation in participating media approach.

Figure 4 and Figure 5 show curves of the mean solid temperature along the channel and its spatial derivative for the 3d model in comparison with calculated curves from the 1d model with varying values of the absorption coefficient. The best agreement for the used configuration is obtained for $\kappa_{rad} = 1300m^{-1}$.

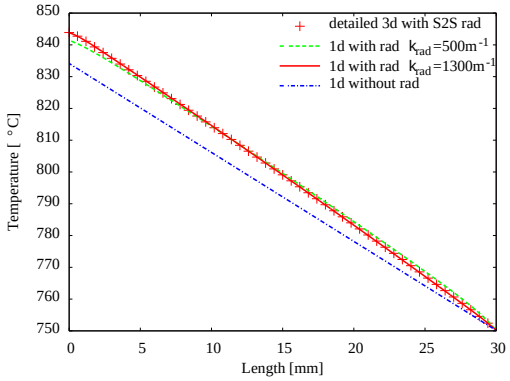

Figure 4. Comparison of temperatures along channel direction for 3d surface-to-surface radiation and the 1d radiation in participating media model for several absorption coefficients.

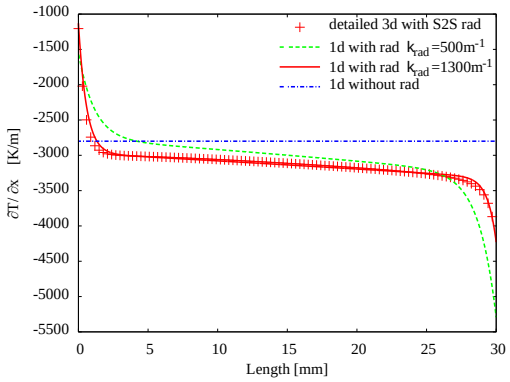

Figure 5. Comparison of temperature gradients along channel direction for 3d surface-to-surface radiation and 1d radiation in participating media model for several absorption coefficients.

5.2 Reforming Unit Model

The main focus of this work is the temperature distribution of the solid honeycomb comparing the cases for heat transport with and without radiation.

In Figure 6 the contour plots of these temperatures are shown. The surface-to-surface radiation tends to equilibrate the temperatures between the participating surfaces which results in increasing temperatures along all boundaries except the honeycomb inlet where the temperature is decreasing. In those regions where the temperature is increasing the fuel gas is pre-

heated because of the net heat flow from boundaries to the free-flow domain. Inside of the honeycomb the gas temperature distributions are different from each other but in the same range.

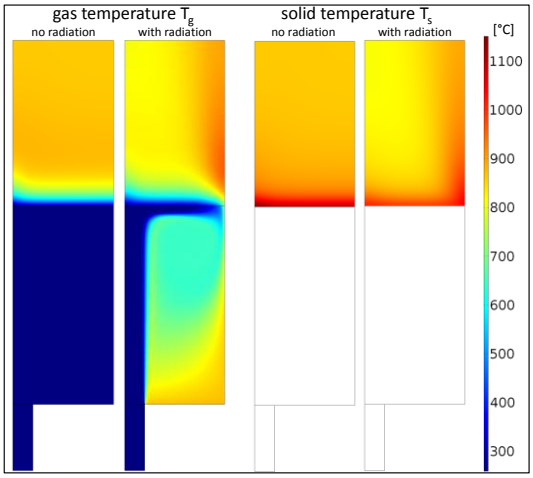

Figure 6. Solid and gas temperature distribution of the reformer unit model with and without radiation.

The contours of the solid temperature are more interesting and of practical relevance. In case of radiation, solid temperatures are nearly everywhere lower than in the pure heat conduction and convection case. The total decrease of the maximal temperature of around 100K is better depicted in Figure 7 where the cross section averaged temperatures for gas and solid are plotted. Radial temperature variations are shown in Figure 8 where for axial lines at $r = 0mm$ and $r = 15mm$ the solid and gas temperatures are plotted.

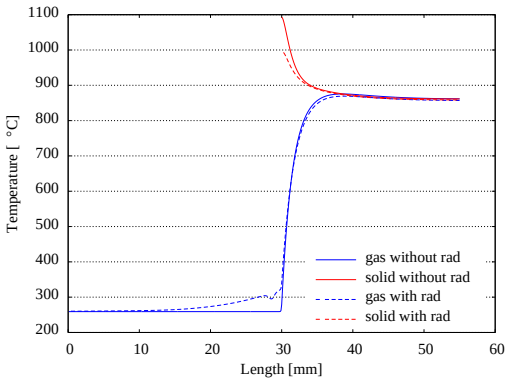

Figure 7. Mean solid and gas temperature along the main gas flow direction with and without radiation.

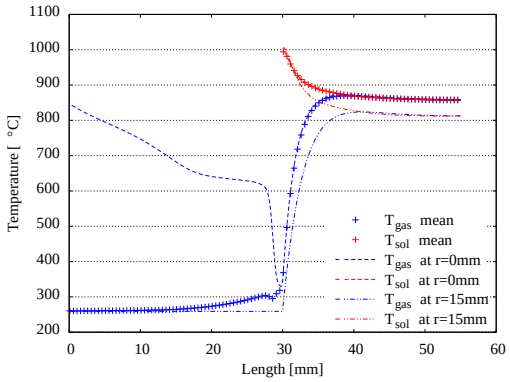

Figure 8. Mean solid and gas temperature along the main gas flow direction in comparison with axial lines at different radial positions.

6. Conclusion and Outlook

Within this work an axis symmetric reformer unit model was developed including mass transport in free-flow and porous regions, most simplified chemical reactions and heat transport including convection, conduction and radiation. The catalytic honeycomb was considered as a porous media with anisotropic properties allowing an axisymmetric implementation.

A developed 1d sub model for radiation in participating media along a channel structure showed good agreement with a detailed 3d surface-to-surface radiation channel model.

The derived reformer unit model showed a significant influence of the temperature distribution on heat transfer by radiation as expected.

The presented model only considers only constant material properties but temperature dependent ones are in progress. The very simple reaction approach will be exchanged by more convenient ones and gas phase chemistry should also be included in further work.