Thermal Characterization of a Chemical Reactor Coupling Comsol and ModeFrontier

N. Pacheco[1,2], D. Pavone*[,1], E. Schaer[2], J.L. Houzelot[2], K. Surla[1]
[1]IFP-Energies nouvelles, [2]LRGP-ENSIC
*Corresponding author: Rond-point de l'échangeur de Solaize, BP3, 69360 Solaize, didier.pavone@ifpenergiesnouvelles.fr

Abstract: For Hydrogen production purpose from bio-ethanol, IFP Energies nouvelles set up a pilot reactor that can work at high temperature (1000°C) and high pressure (20 bar). Experiments show that this reactor has a specific thermal behaviour that should be modelled in addition to chemical and hydrodynamics behaviour to understand and optimize hydrogen production.
The multiphysics simulator is defined in Comsol and the thermal behaviour is matched on specific thermal experiments designed for the purpose. Thermal parameters that characterize the reactor are estimated coupling ModeFrontier to the reactor simulator. Efficient parameter estimations could be obtained using genetic algorithms leading to an accurate thermal simulator.
We present the simulator and the way it is linked with the optimizer.

Keywords: Heat Loss, Estimation of parameters, modeFrontier.

1. Introduction

The hydrogen is nowadays considered as a promising energy source for the future. It appears as an interesting way to limit the use of fossil fuels and it could allow to reduce the emissions of greenhouse gas. However, the hydrogen is not a primary source of energy and therefore it must be produced. IFP Energies nouvelles develops for several years economic and clean processes of hydrogen production such as the Autothermal Reforming (ATR) using bio-ethanol as feed. However, to reach a high hydrogen level, ATR should work at high temperatures such as 600°C.

To understand and optimize hydrogen production with ATR, IFP Energies nouvelles developed a pilot reactor already presented at 2008 Comsol conference in Hanover[1]. This pilot is very close to actual units as it can reach 1000°C and 20 bar for operating conditions. However, while running the experiments, we found that the reactor exhibits heat losses. Generally, in such a research work, people assume that (i) the heat loss are negligible and the reactor can be taken as adiabatic, or (ii) the heat loss are huge enough that an isothermal approach is correct. In our case, we measured a huge increase in the temperature close to the gas entrance and then a smooth decreasing temperature profile to the exit. It occurs that the reactor is neither adiabatic nor isotherm and the actual thermal behavior of the reactor should be taken into account together with the complete chemical reactor simulator to analyze experiments.

From simulation point of view, nowadays we have tools that can help to model multiphysics process. But simulators come with parameters that are most of the time unknown and they should be estimated with one optimization software. The same holds true for process optimization itself. At IFP Energies nouvelles we are using modeFrontier as flexible and powerful optimizer. We show here how we link the simulation in Comsol with the optimization in modeFrontier to get a representative simulator of our actual pilot reactor.

This paper presents

1. The thermal experiments performed to characterize the reactor,
2. The thermal simulator of the ATR reactor
3. The coupling between the simulator and the optimizer.

2. Heat loss experiments

A typical heat loss experiment consists into heating the reactor furnaces to a set temperature without gas flow in the reactor. Once at steady state, the heating furnaces are turned off and the reactor natural cooling is followed each 10s by 11 thermocouples (Figure 1). The experiment stops when all temperatures are below 473K (200°C).

We performed six experiments with six different initial temperature values at 400°C, 500°C, 600°C, 700°C, 800°C and 900°C.

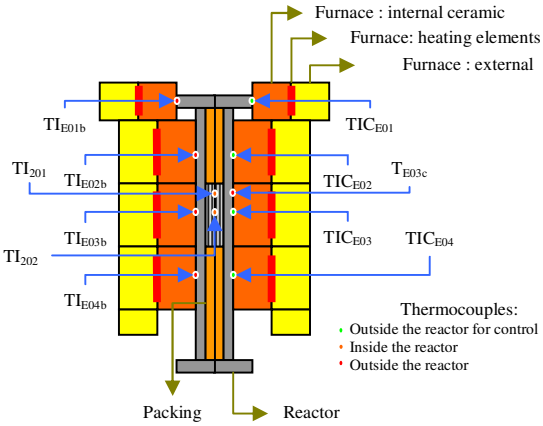

Figure 1: Scheme of furnaces, reactor and thermocouple locations

Figure 1 presents the location of the 11 thermocouples (in red and green). Some thermocouples are facing to check the heating symmetry of the reactor.

3. Numerical simulator for thermal behaviour

A numerical simulator devoted to heat loss analysis has been built in Comsol Multiphysics 3.5. The 3.5a version is not given with the "script" and hence can not be linked with ModeFrontier. The procedure to link Comsol and modeFrontier is explained below.

The partial differential equation for the energy balance (1) is solved by finite element method, in 2D axial symmetry geometry (Figure 2).

$$\rho_i Cp_i \frac{\partial T}{\partial t} = \nabla \cdot (\lambda_i \nabla T) \qquad (1)$$

Where ρ is the density (kg/m^3), Cp is the heat capacity (J/kg.K), λ is the thermal conductivity (W/m.s), T is temperature (K) and t the time (s).

Figure 2 presents the 2D axial symmetry geometry based on the equipment actual dimensions. Boundary conditions are presented as well.

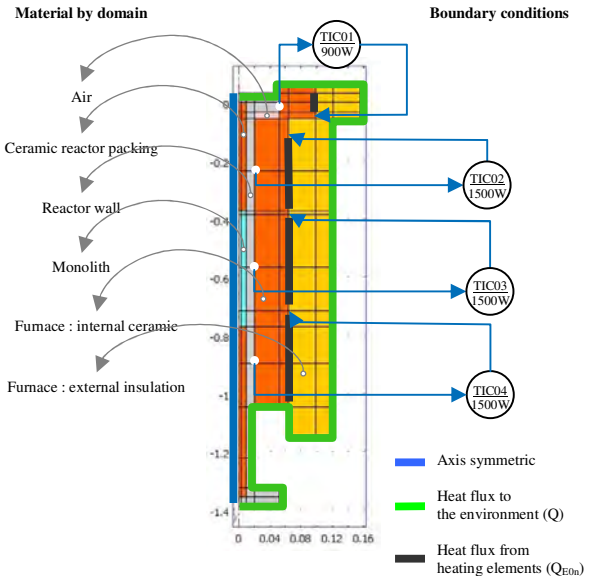

Figure 2: Simulated furnaces and reactor

On figure 2, six different domains are identified. Each domain has its own physical properties. Some are known but the ones for the furnace should be estimated matching experiments. There are three unknown parameters for each furnace domains (internal ceramic and external insulation) i.e. ρ, Cp and λ. But actually, they can be reduced to two merging ρ and Cp into (ρ Cp).

The heat flux towards the environment (in green) is calculated using equation (2).

$$Q = h(T - T_\infty) \qquad (2)$$

Where Q is the heat flux for the environment (W/m^2), h is the global heat transfer coefficient (W/m^{2}K) and T_∞ is the environment temperature (K). The global heat transfer coefficient h is calculated from literature correlations [2].

The heat flux from the heating elements is calculated simulating the proportional and integral controller (PID) behavior (3). The furnace control system is simulated to correctly determine the initial conditions at steady state, before transient cooling.

$$QE0n = [0 < Kp(TICn - TICn_{SP}) + \\ + \frac{1}{Ki}\int_0^t (TICn - TICn_{SP})dt < 1]P_{E0n}^{\max} \quad (3)$$

Where Q_{E0n} is the heat flux (W/m^2) from the heating element n, $P_{E0n}^{\max}$ is the maximum heating flux (W/m^2) that could be delivered by the heating element n, $TIC0n$ is the calculated temperature (K) at the same position as for the experimental thermocouple $TIC0n$, $TIC0n_{SP}$ is the set point temperature (K) for furnace n, t is the time (s), Kp is the proportional constant equal to -0.1K^{-1}, and Ki is the integral constant equal to -10^{6}K/s.

The simulation is transient and follows the experiments: the temperature set point is defined for each furnace at steady state; the PID control achieves the steady state and once at steady state the furnaces are turned off.

4. Estimation of furnace physical properties

The furnace physical properties are not known beforehand therefore they should be estimated using the experimental data and the numerical simulator described before. Four parameters are estimated using modeFrontier (4.1.1 by MOGAII methods[3]) : the product $\rho.Cp$ and the conductivity λ for each furnace section (internal ceramic and external insulation). The objective function to be minimized was the squared error sum between simulation and experiment.

For each of the six experiments, four constant parameters were estimated. It occurs that the parameters are functions of the temperature (Figure 3 and Figure 4). Figure 3 shows the estimated thermal conductivities for both sides of the furnaces (inside and insulation) as temperature dependent.

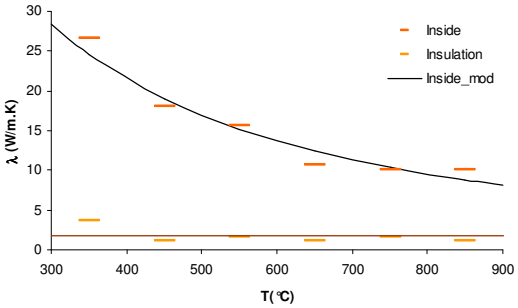

Figure 3: Thermal conductivities of furnace inside and insulation

The thermal conductivity of the inside ceramic is much higher than the insulation and it reduces with the temperature while for the external insulation the thermal conductivity is almost constant. Hence, for the insulation ceramic we consider the mean value of 1.72W/m.K but for the inside ceramic, the thermal conductivity depends on the temperature through (4), with a squared correlation factor of 0.94.

$$\lambda_{inside} = 1.76 \times 10^6 \, T^{-1.74} \quad (4)$$

Where λ_{inside} is the estimated thermal conductivity (W/m.s) for the inside ceramic and T is the temperature (K).

Figure 4 shows the estimated $\rho.Cp$ for both sides of the furnaces as a temperature dependent.

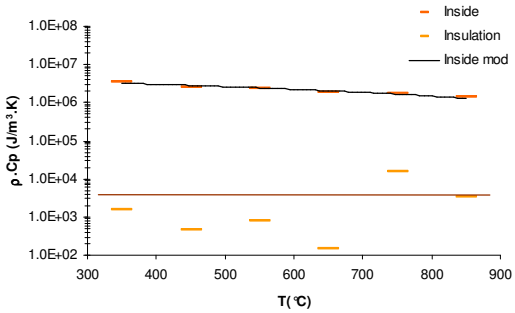

Figure 4: $\rho.Cp$ of internal ceramic side and external insulation

The inside ceramic has a very high $\rho.Cp$ compared to the external insulation. Figure 4 also shows a great uncertainty for the product $\rho.Cp_{insulation}$. For the insulation

material, the mean value of $\rho.Cp_{insulation}$ is taken (3.69×10^3 J/m^{3}K) and for the inside ceramic a linear correlation (5) is taken with a squared correlation coefficient of 0.94.

$$\rho Cp_{inside} = -3.92\times10^3 T + 5.68\times10^6 \qquad (5)$$

Where ρCp_{inside} (W/m.s) is the estimated correlation for the inside ceramic and T is the temperature (K).

Introducing the estimated data and correlations ((4) and (5)) in the simulator, we ran heat loss simulations and compared the simulator results to the experimental data for numerical validation in Figure 5.

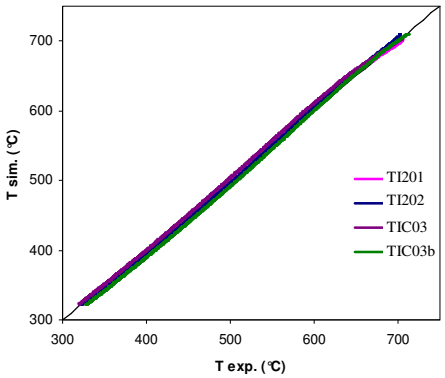

Figure 5: Experimental versus simulated comparison for the experiment at 700°C (973K)

At steady state, the axial profile of temperatures was measured and used as comparison with the simulated data (Figure 6). These data were not used to estimate the parameters.

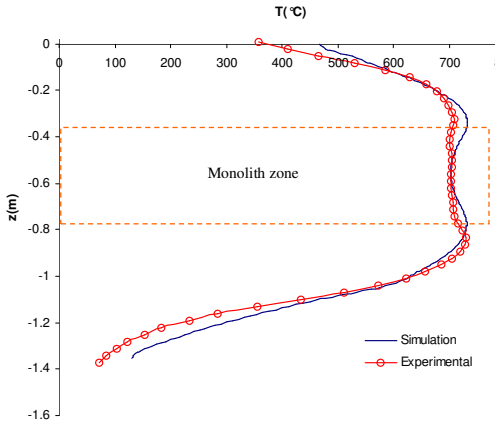

Figure 6: Experimental results with simulation comparison, at steady state for 700°C (973K)

Figure 6 shows a rather good agreement between experimental results and the simulation. The calculated temperature map and the heat flux vectors are shown in Figure 7, for the same steady state as shown in Figure 6.

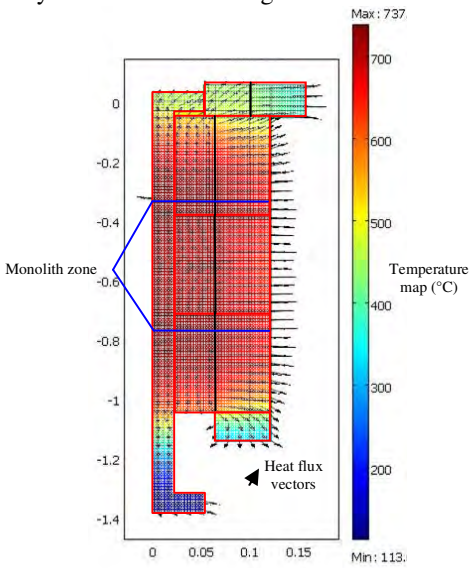

Figure 7: Calculated temperature map and heat flux vectors for steady state at 700°C

As a conclusion we can say that the estimated parameters are correctly estimated and that the thermal behaviour of the reactor can be taken into account for chemical reaction simulations that will follow this work.

5. Optimization process using Comsol 3.5 linked to modeFrontier 4.1

A workflow is defined on modeFrontier. The workflow is constituted of process flow and dataflow. The process flow describes the sequences of actions and the data flow describes what pieces of data should be moved from one step to another. Figure 8 shows the modeFrontier workflow for parameter estimation using ATR pilot reactor model.

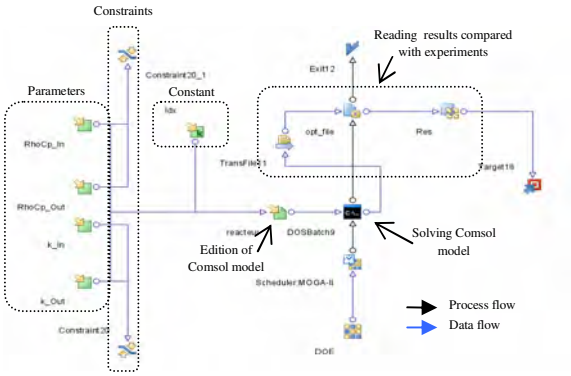

Figure 8: Workflow on modeFrontier for parameters estimation using Comsol

The scheduler defines the set of parameters that will be used in the numerical simulator, by the MOGA-II genetic algorithm. The Comsol model is an "m-file" created by Comsol and edited later by the user for modeFrontier application. The structure of the m-file generated by Comsol and the modifications that need to be done for modeFrontier are shown in Figure 9.

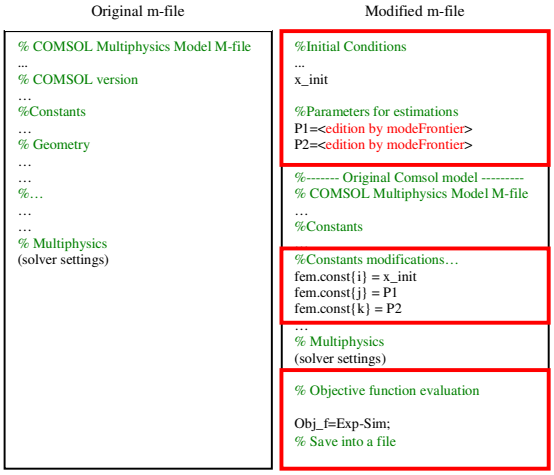

Figure 9: Structure of m-file generated by Comsol (left) and the modified one for modeFrontier

Three main modifications need to be done to the original m-file: definition of initial conditions of experiments (*x_init*) and parameters for estimation (P_i); modifications of constants in original comsol model; and finally evaluation of objective function related to the experimental observations.

ModeFrontier modifies the parameters *Pi* by changing its value in the m-file, then Comsol batch run the modified m-file producing a results file, which is read by modefrontier for evaluation of the target.

Through this methodology, the numerical simulator developed in comsol is linked to modeFrontier for model parameter estimation.

6. Conclusion

The thermal behaviour of the ATR pilot reactor is modelled using Comsol 3.5. This numerical simulator has four unknowns that are successful estimated using the optimizer modeFrontier 4.1. The parameter estimation is done with experimental data. Hence, a representative numerical simulator of the ATR pilot reactor is available to help the analysis of catalytic experiments.

8. References

1. Pacheco, Simulation of Surface Chemical Reactions in a Monolith Channel for Hydrogen Production, Comsol Multiphysics Conference (2008)
2. Greth - MTI, V3.0, (2004)
3. modeFrontier 4.0: Users manual, 4.0, (2009)

9. Acknowledgements

The authors thank ANRT for the financial support. Nuno Pacheco is grateful to IFP Energies nouvelles and ENSIC for providing a doctoral grant.