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Modeling of Transport and Reaction in a Catalytic Bed Using a Catalyst Particle Model.

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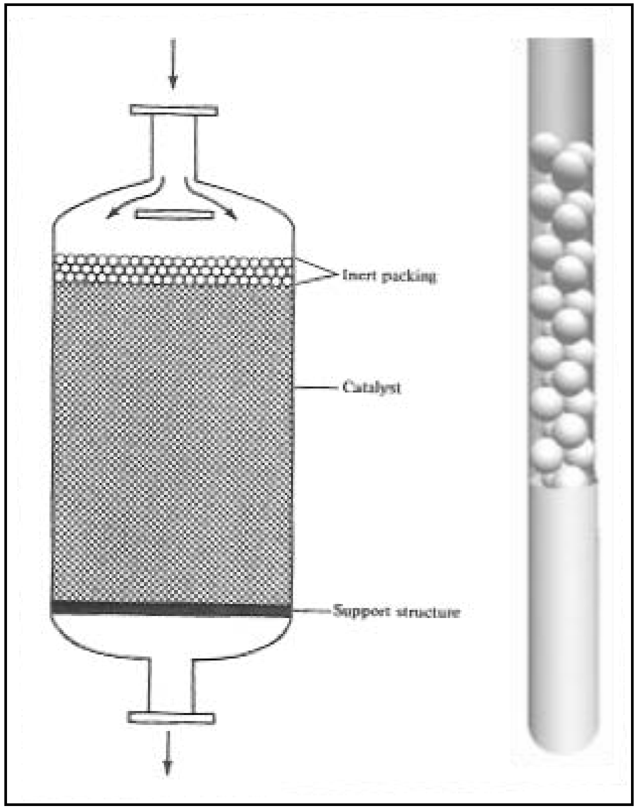
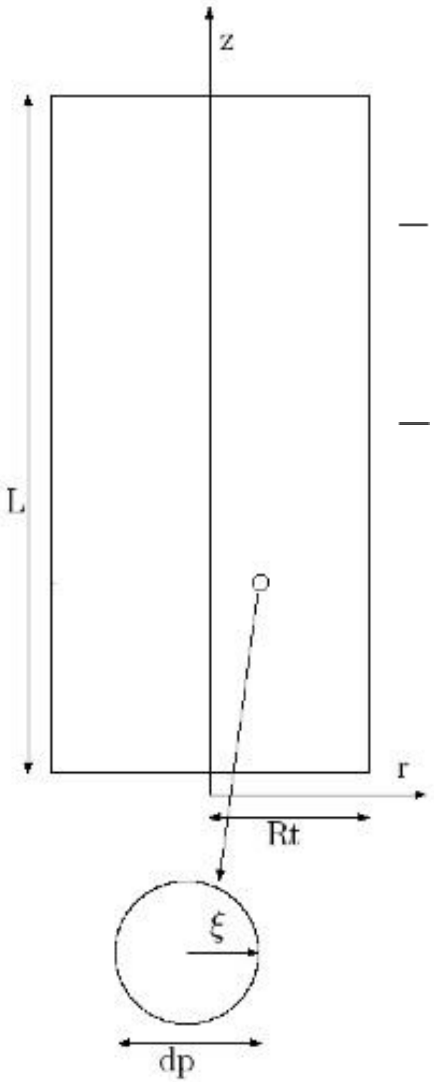
Background



- Packed bed reactors: CO conversion, MSR reaction, Benzene conversion to cyclohexane...
- Characterized by N=ratio between bed and particle diameter,
- Usual model: pseudo-heterogeneous, effectiveness factor
  → High N
- Our model: pseudo-heterogeneous, diffusion in the solid particles.

Our model (1)



- Fluid equations:

$$-\frac{\partial C_i'}{\partial z'} + \frac{1}{Pe_{m_{r,i}}}\left(\frac{\partial^2 C_i'}{\partial r'^2} + \frac{1}{r'}\frac{\partial C_i'}{\partial r'}\right) + \frac{1}{Pe_{m_{a,i}}}\frac{\partial^2 C_i'}{\partial z'^2} = St_{m,i}(C_i' - C_{i,s}'^{s})$$

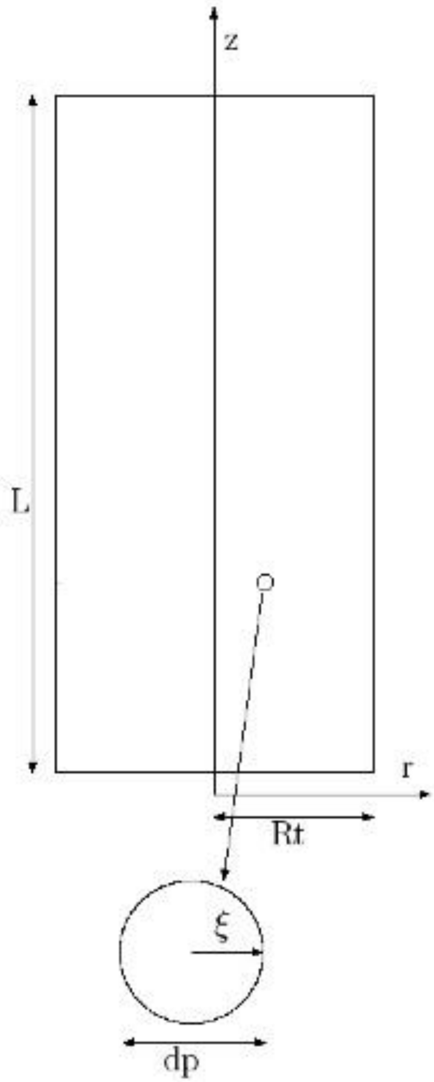
$$-\frac{\partial T'}{\partial z'} + \frac{1}{Pe_{h_r}}\left(\frac{\partial^2 T'}{\partial r'^2} + \frac{1}{r'}\frac{\partial T'}{\partial r'}\right) + \frac{1}{Pe_{h_a}}\frac{\partial^2 T'}{\partial z'^2} = St_h(T' - T_s'^{s})$$

- Solid equations:

$$-\frac{\partial^2 C_{s,i}'}{\partial \xi'^2} = \frac{2}{\xi'}\frac{\partial C_{s,i}'}{\partial \xi'} + \phi^2 rate$$

$$-\frac{\partial^2 T_s'}{\partial \xi'^2} = \frac{2}{\xi'}\frac{\partial T_s'}{\partial \xi'} + \beta\phi^2 rate$$

Our model (2)



- Boundary conditions:
  - Fluid phase:

$$z' = 0 \qquad -\frac{\partial C_i'}{\partial z'} = Pe_{m_{a,i}}(C_i' - C_{i,in}')$$

$$-\frac{\partial T'}{\partial z'} = Pe_{h_a}(T' - T_{in}')$$

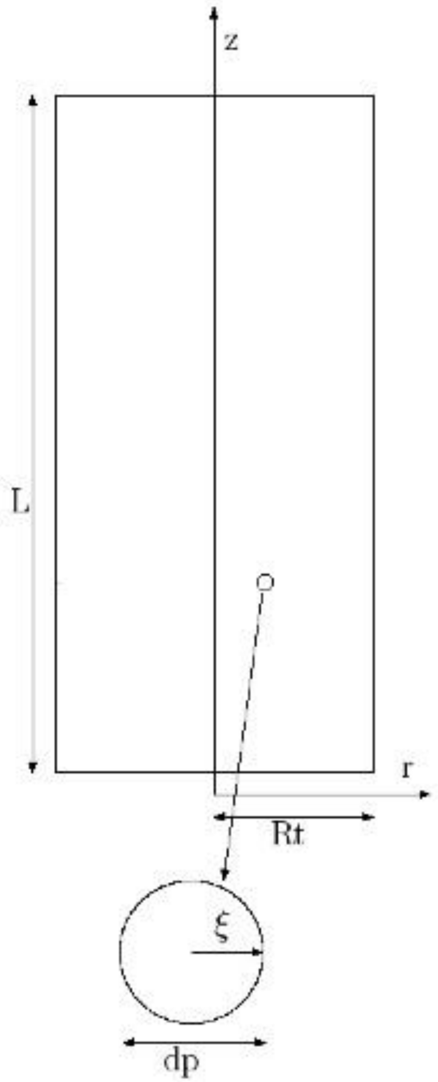
$z' = \dfrac{L}{Rt}$ Convective fluxes for C and T

$r' = 0$ Axis symmetry for C and T

$$r' = 1 \qquad \frac{\partial C_i'}{\partial r'} = 0 \quad (insulation)$$

$$-\frac{\partial T'}{\partial r'} = Bi_w(T' - T_w')$$

Our model (3)



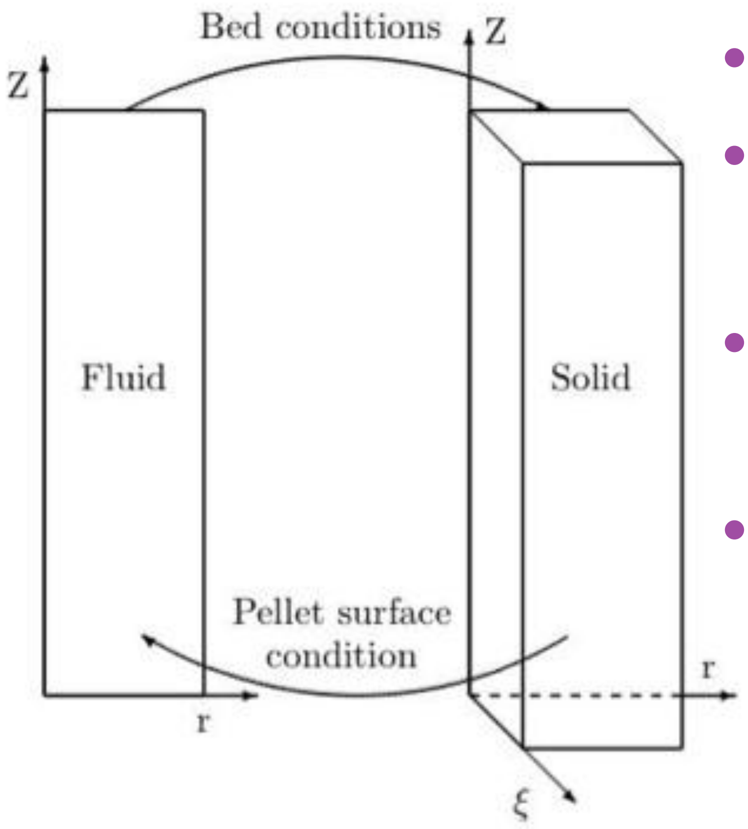
- Boundary conditions:
  - Solid phase:

$$\xi' = 1 \qquad -\frac{\partial C'_{s,i}}{\partial \xi'} = Sh_{s,i}(C'_{s,i} - 1)$$

$$-\frac{\partial T'_s}{\partial \xi'} = Bi_s(T'_s - 1)$$

$\xi' = 0$ Axis symmetry

Implementation in COMSOL© 3.5



- Chemical eng. module
- Fluid equations: 2D Convection conduction/diffusion
- Solid equations: 3D Conduction/diffusion
- Extrusion coupling variables from one geometry to the other.

Test case - Statement

- First case: CO conversion over copper-chromite catalyst. N=11.

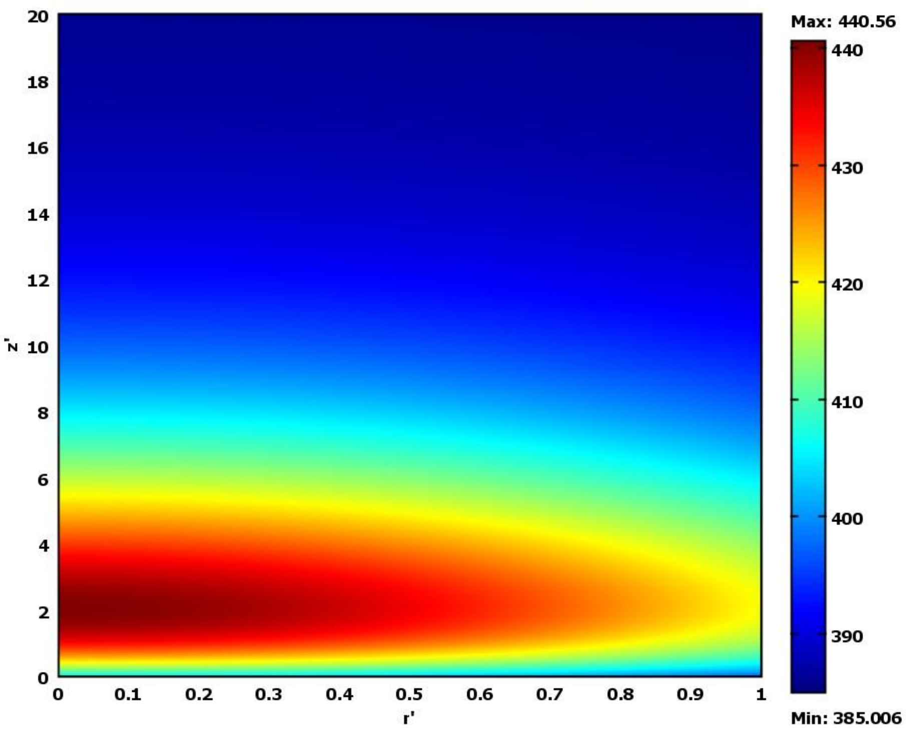
$$CO + \frac{1}{2}O_2 \longrightarrow CO_2$$

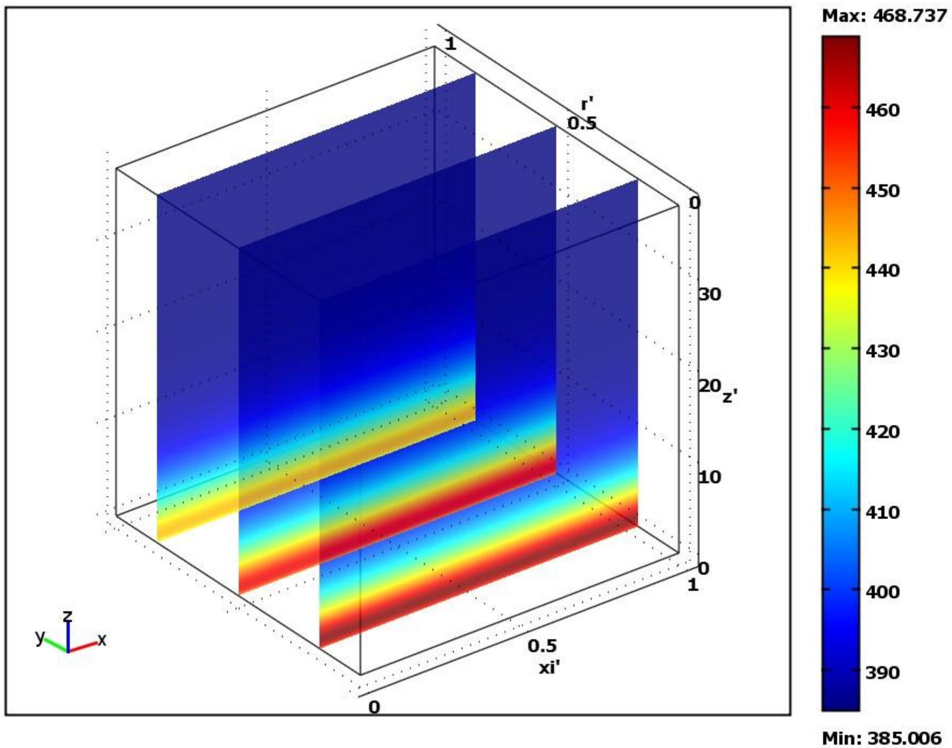
- CO inlet concentration of 0.5 mol/m^3,
- O_2 considered constant (30.6 mol/m^3) in the fluid and solid,
- Constant wall temperature of 385K.

Test case – Results (1)

Temperature profile – Fluid (K)

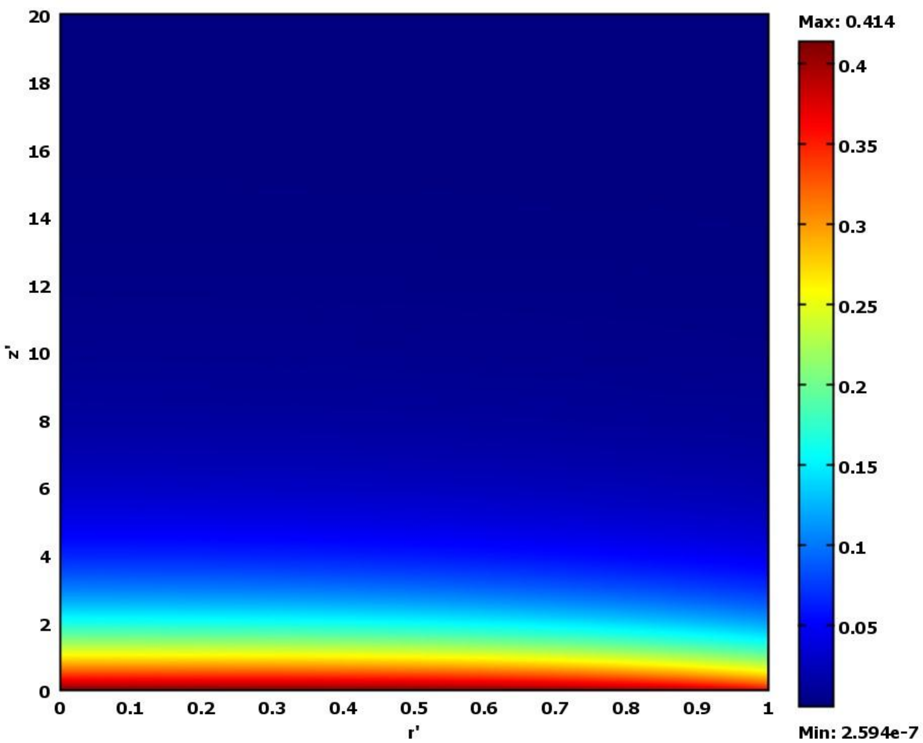
Temperature profile – Solid (K)



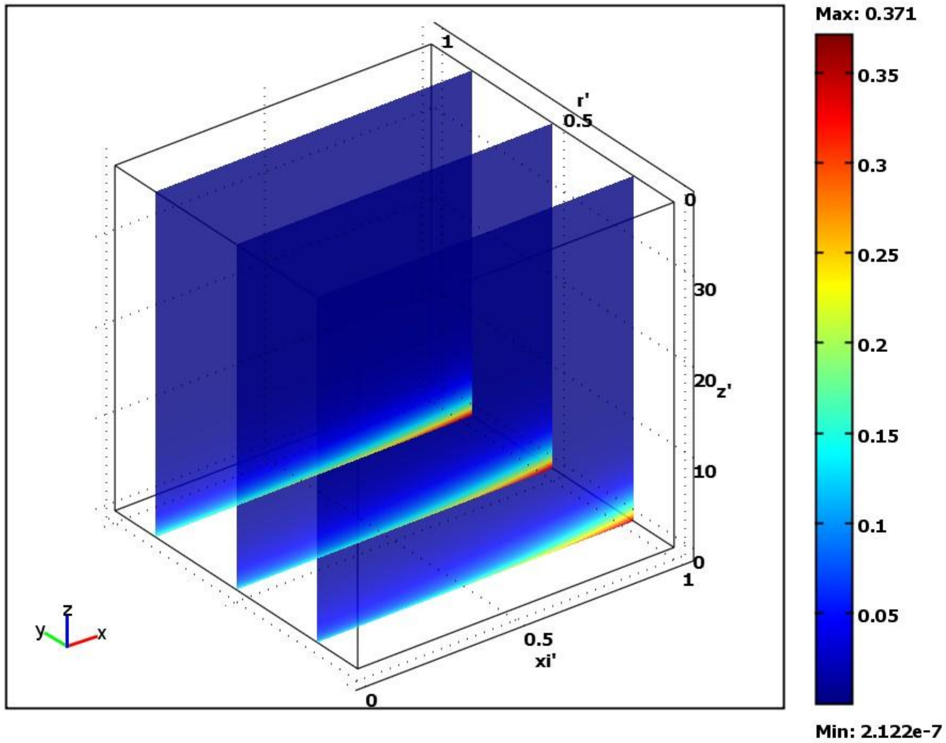


Test case – Results (2)

CO concentration profile – Fluid (mol/m³)



CO concentration profile – Solid (mol/m³)



Test case - Comparison

Machac et al. 2006	**Present study**
• Hot spot in the center, at z'= 2 • Hot spot temperature: 475 K • Temperature difference solid/fluid, r'=0.5: 20K • Effective inlet concentration: around 0.45mol/m^3	• Hot spot in the center, at z'= 2.5 • Hot spot temperature: 440 K • Temperature difference solid/fluid, r'=0.5: 43K • Effective inlet concentration: 0.414 mol/m^3

- Similar profiles for both concentration and temperature in the fluid phase.
- Need to get better coefficient for conduction/diffusion in the solid.

Study case - Statement

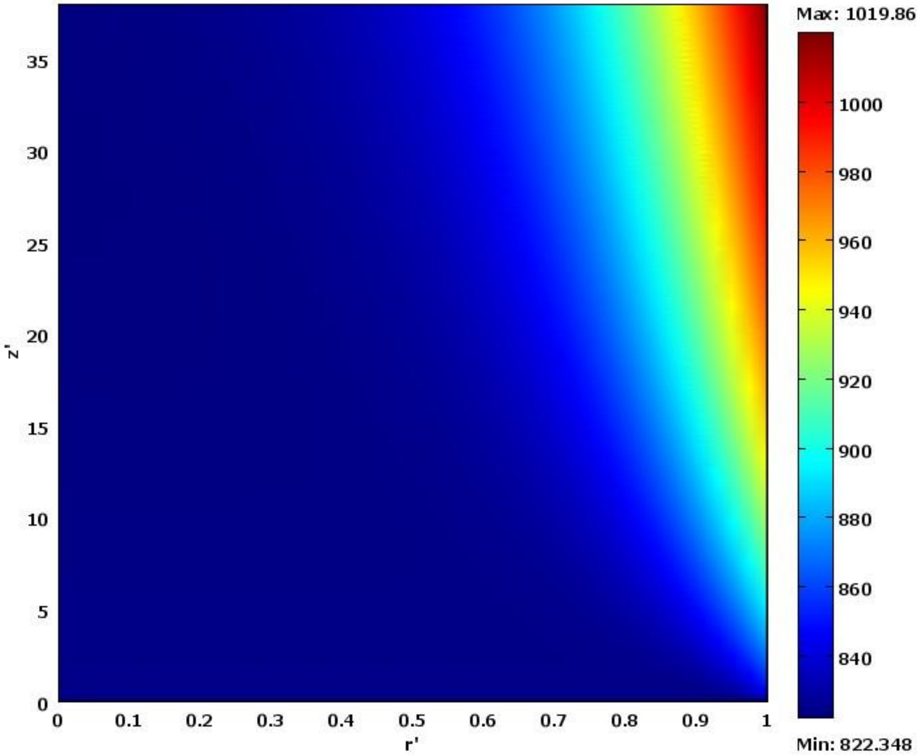
- MSR reactions. N=6

$$CH_4 + H_2O = CO + 3H_2$$
$$CO + H_2O = CO_2 + H_2$$
$$CH_4 + 2H_2O = CO_2 + 4H_2$$

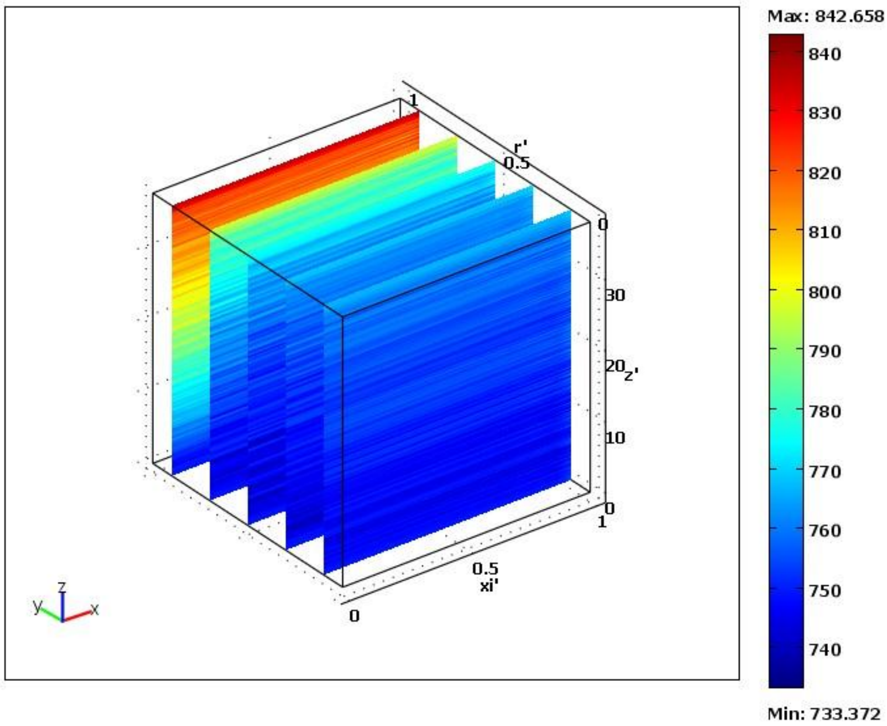
- Reaction rates from Hou et Hughes. *The kinetics of methane steam reforming over a Ni/[alpha]-Al2O catalyst*. Chemical Engineering Journal (2001)
- Data from simulations by Johnson Matthey.

Test case – Results (1)

Temperature profile – Fluid (K)

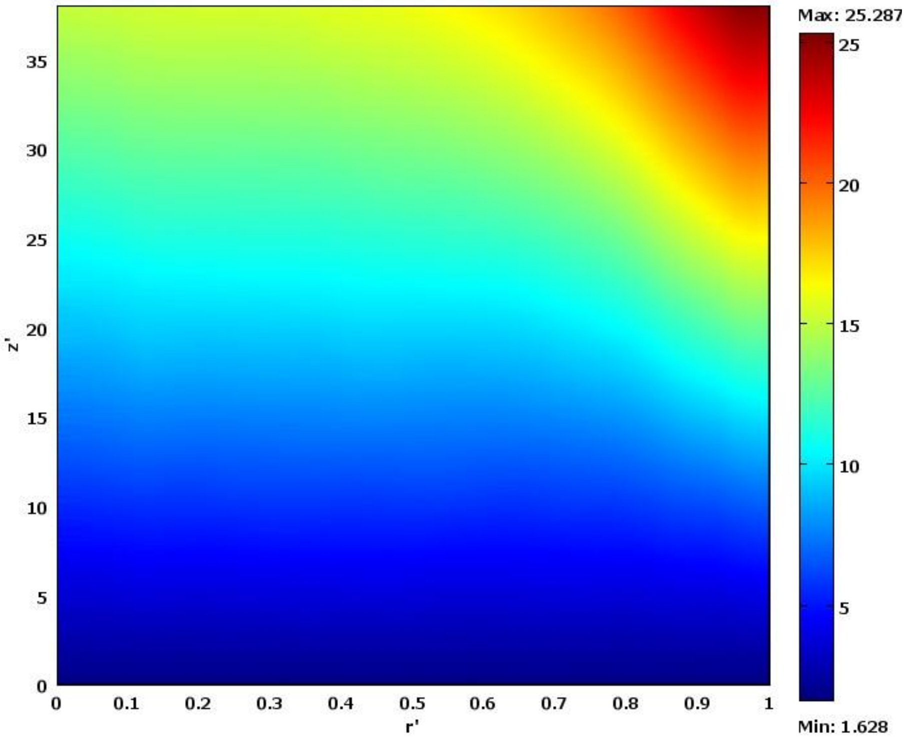


Temperature profile – Solid (K)

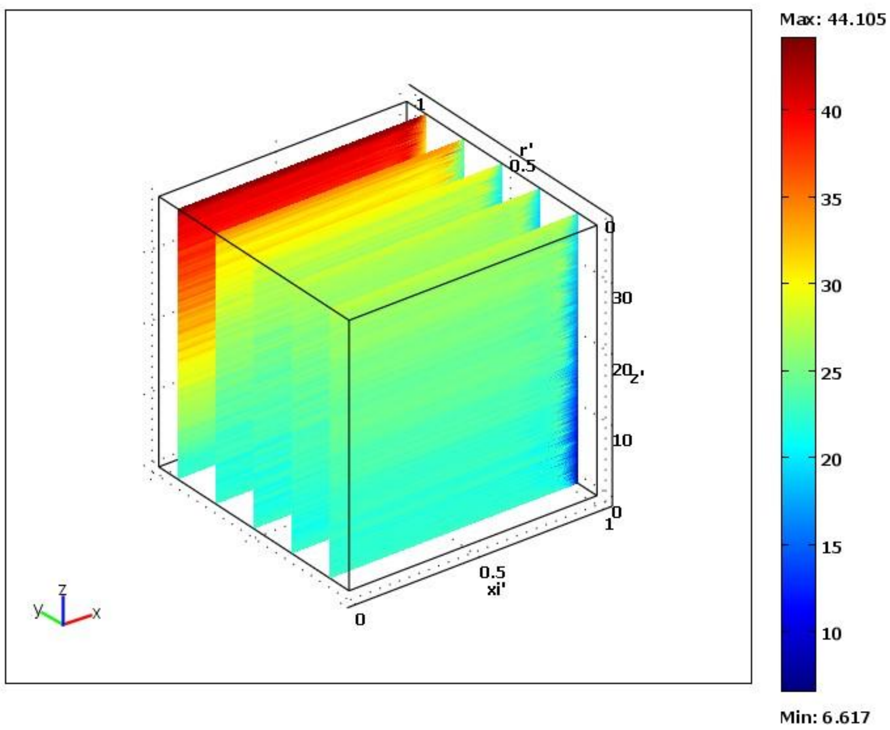


Test case – Results (2)

H_2 concentration profile – Fluid (mol/m³)



H_2 concentration profile – Solid (mol/m³)



Conclusion

- Good agreement in the test case with literature,
- Model applicable to more complex reactions,
- Need to reevaluate the heat and mass transfer coefficient for MSR reaction, for both fluid and solid phase.

Acknowledgments

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- COMSOL