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CFD multiscale simulation of multiple gas-solid reactions in the industrial ironmaking process

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CFD MULTISCALE SIMULATION OF MULTIPLE GAS-SOLID REACTIONS IN THE INDUSTRIAL IRONMAKING PROCESS

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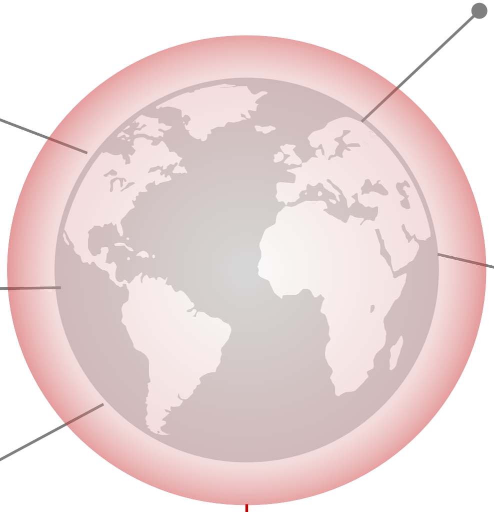
Agosto 2024

Direct Reduction Of Iron Ore (DRI)



~130 million tons of iron
is the world production in 2022.
(MIDREX, 2024)

High energy consumption
is decreased compared to blast furnace
(IEAGHG, 2018)

Smaller units are more flexible
(Béchara et al., 2018)

Reduction in CO_2 emissions
40 to 50% compared to blast furnace (IEAGHG, 2018)
Zero emission using only H_2

Substitution for green H_2
via electrolysis
(Patisson and Mirgaux, 2020)

climate change

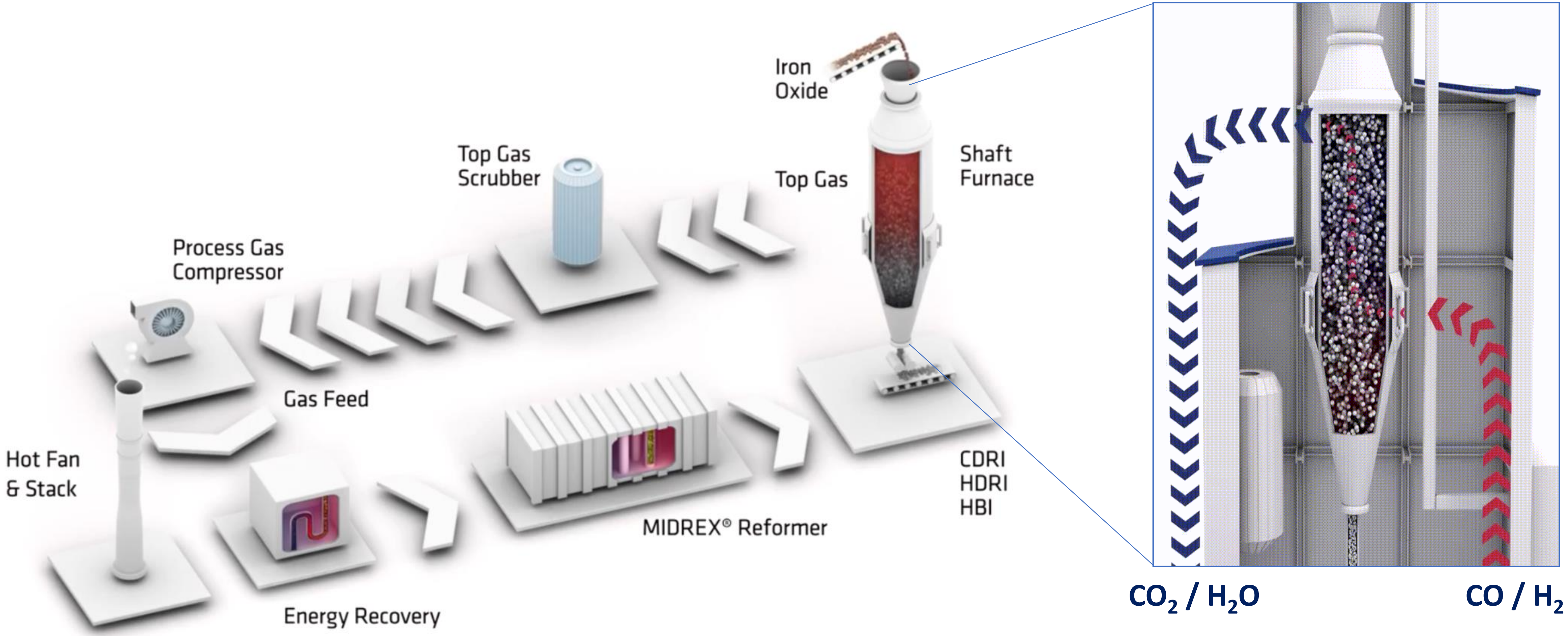
Steelmaking (1/3 of industrial CO_2 emissions)
(IPCC, 2014)

Paris Agreement
European Green Deal

- Limit global warming to 1.5 °C by 2050
- 55% in CO_2 emission until 2030 and carbon neutrality by 2050

Direct Reduction of Iron Ore (DRI)

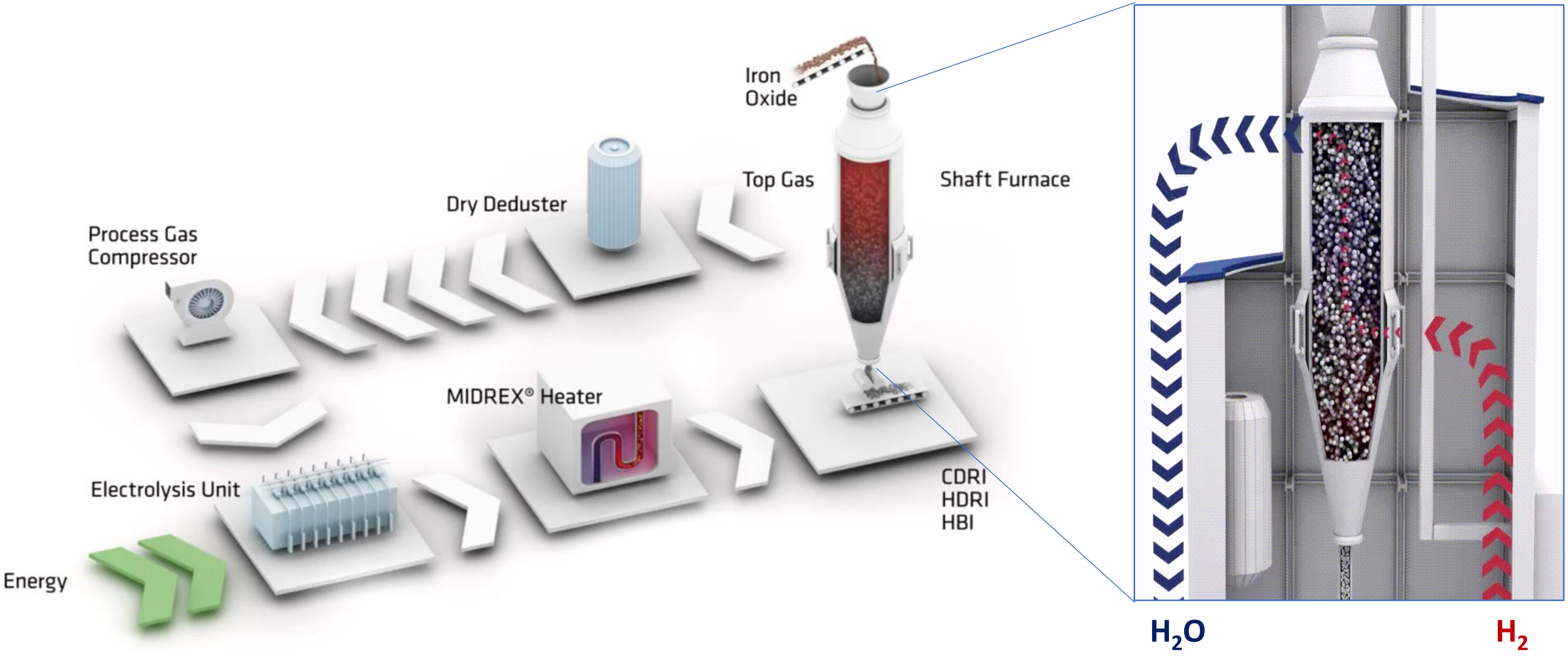
MIDREX Process



MIDREX (2021)

Direct Reduction of Iron Ore (DRI)

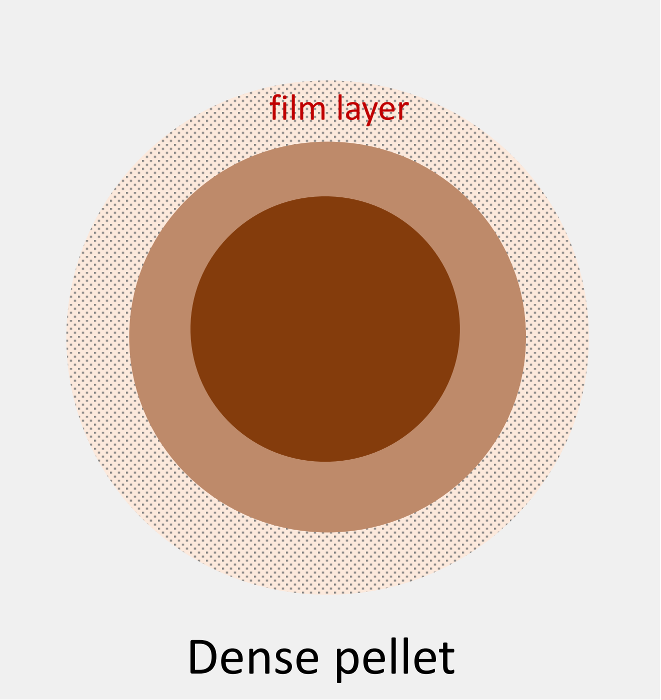
Hydrogen Direct Reduction (H-DR) Process



MIDREX (2021)

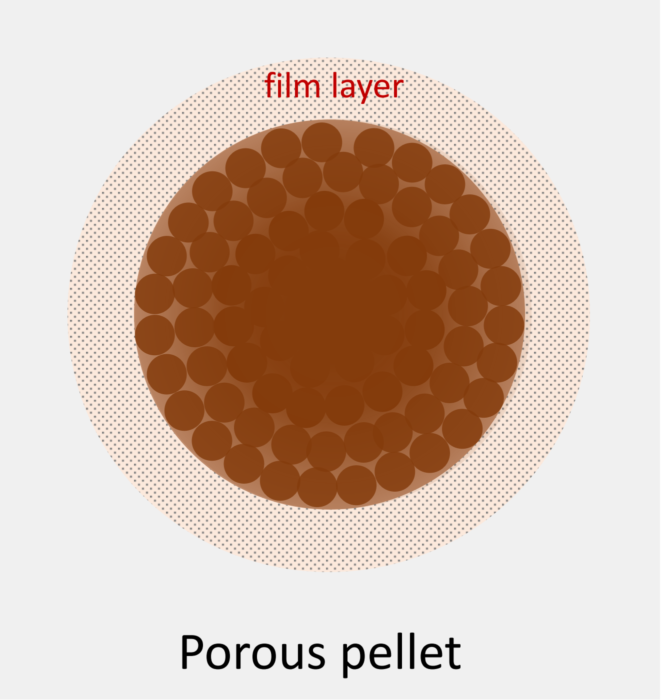
Non-catalytic gas-solid reactions models

Shrinking Core Model (SCM)



Dense pellet

Grain Model (GM)



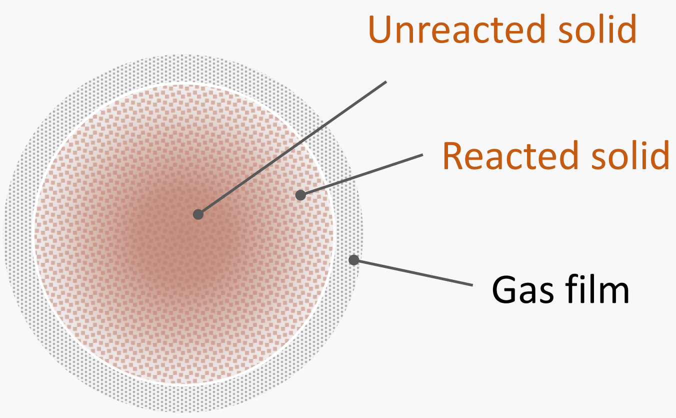
Porous pellet

Homogeneous Model

High porous pellet

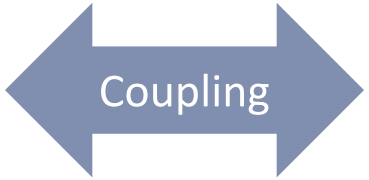
Heterogeneous ⟷ **Mathematical models** ⟷ Homogeneous

Non-catalytic gas-solid reactions models

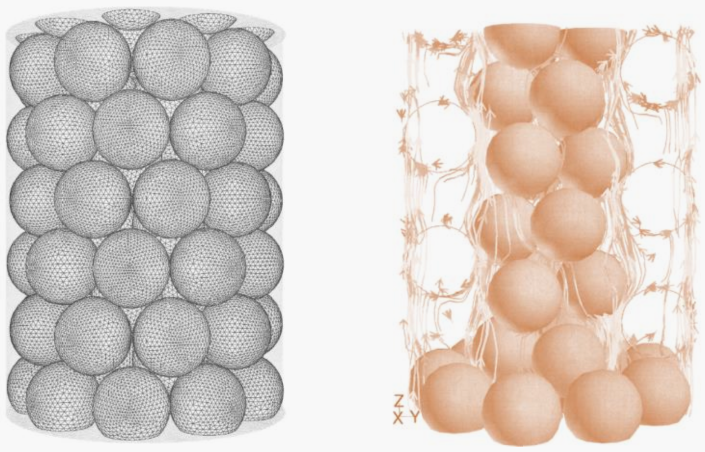


Particle (inside modeling)

- ✓ Mass and heat transport mechanisms
- ✓ Structural changes over time
- ✓ Physical properties of the systems changes



Multi-scale approach



Moving / Packed Bed

- ✓ Mass and heat transport mechanism
- ✓ Fluid flow between particles
- ✓ Random arrangement of particles

OBJECTIVES

Develop a multiscale mathematical model to represent non-catalytic gas-solid reactions applied to the direct reduction of iron ore process

1. Coupling the transport mechanisms in the moving bed reactor with the transport mechanisms and reaction kinetics that occur inside the pellet.
2. Validate the models with industrial data.
3. Predictions for the hydrogen direct reduction process (H-DR).

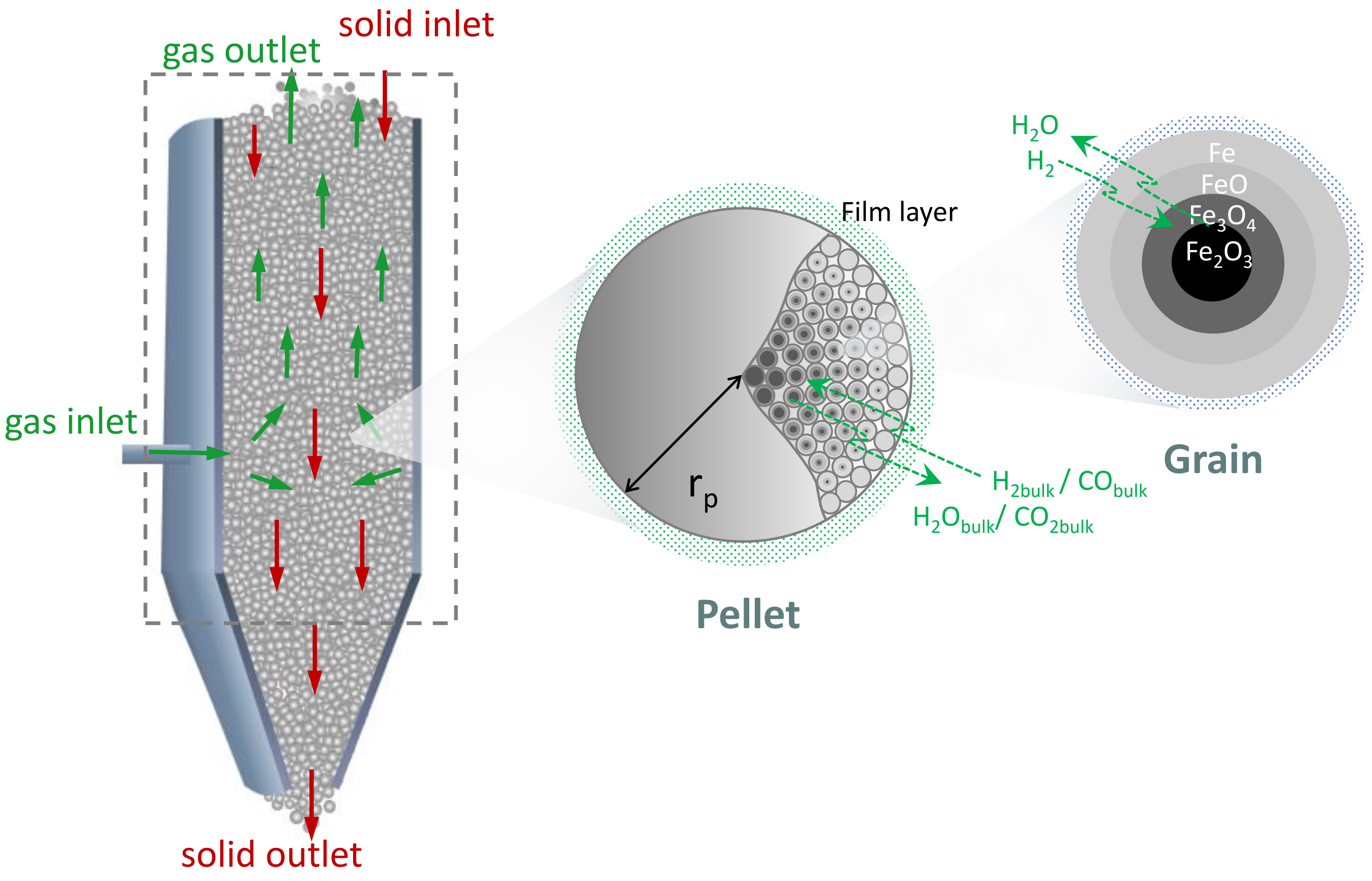
Multiscale Mathematical Modeling

✓ Main assumptions:

- steady state reactor operation
- Ideal gas mixture
- Spherical pellet and grain
- Pellet and grain size constant
- Porosity change
- Non-isothermal pellets
- Grain model for pellet

✓ Finite element method

- Comsol Multiphysics 6.0

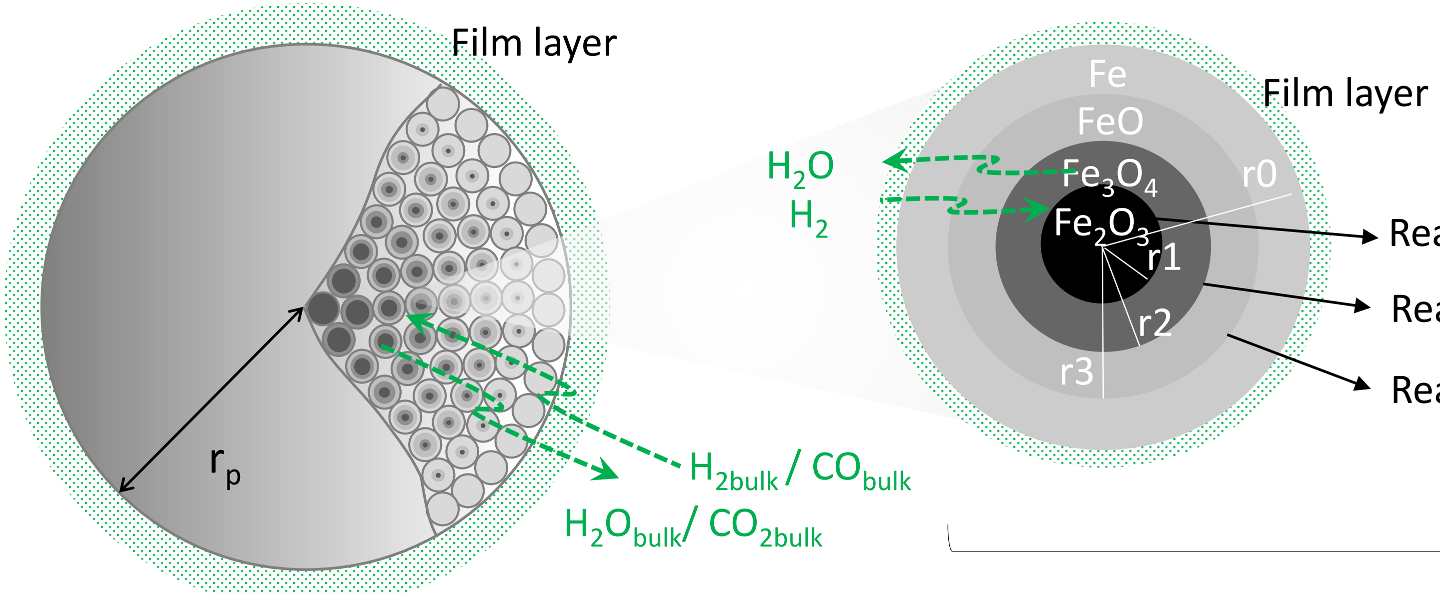


Moving-bed reactor

Multiscale Mathematical Modeling

✓ Pellet model (1D)

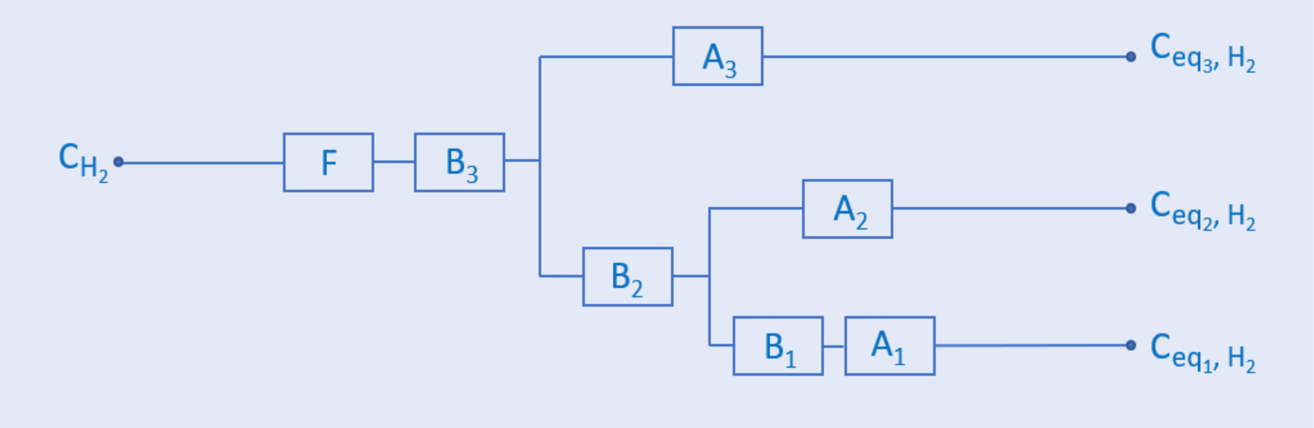
✓ Grain model (1D)



Reaction 1: $3\ Fe_2O_3 + H_2/CO \rightleftharpoons 2\ Fe_3O_4 + H_2O/CO_2$

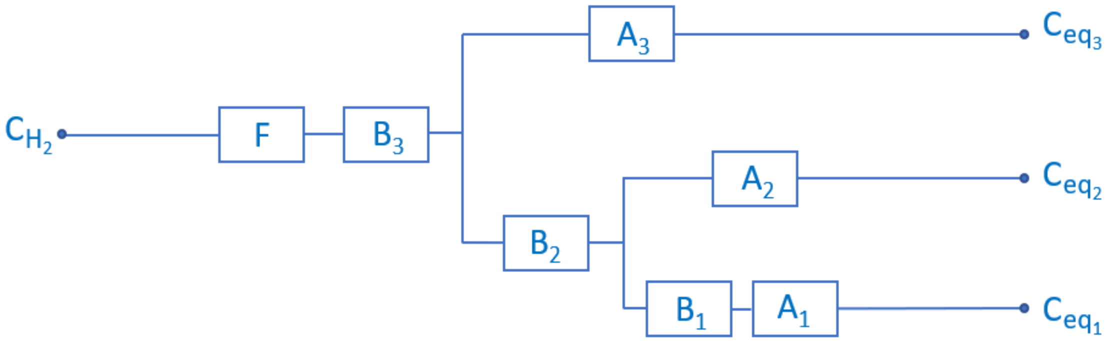
Reaction 2: $Fe_3O_4 + H_2/CO \rightleftharpoons 3\ FeO + H_2O/CO_2$

Reaction 3: $FeO + H_2/CO \rightleftharpoons Fe + H_2O/CO_2$



Multiscale Mathematical Modeling

✓ **Grain model**



A_n: Chemical reaction
B_n: Diffusion
F: Film layer
Ceq: Equilibrium concentration

$$R_{1,i'} = \frac{3}{r_g}\frac{1}{W_{i'}}\{[A_3(A_2+B_2+B_3+F)+(A_2+B_2)\,(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_1,i'}\right) - (B_2(A_3+B_3+F)+A_3\,(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_2,i'}\right) - [A_2(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_2,i'}\right)\}, \qquad i' = \mathrm{H_2 \ or \ CO}$$

$$R_{2,i'} = \frac{3}{r_g}\frac{1}{W_{i'}}\left\{\left[-\left(B_2\,(A_3+B_3+F)+A_3\,(B_3+F)\right)_{i'}\left(C_{p,i'}-C_{eq_1,i'}\right) + [\,(A_1+B_1+B_2)(A_3+B_3+F)+A_3\,(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_2,i'}\right) - [(A_1+B_1)\,(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_3,i'}\right)\right]\right\}, \qquad i' = \mathrm{H_2 \ or \ CO}$$

$$R_{3,i'} = \frac{3}{r_g}\frac{1}{W_{i'}}\,\{-[A_2(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_1,i'}\right) - [(A_1+B_1)\,(B_3+F)]_{i'}\left(C_{p,i'}-C_{eq_2,i'}\right) + [(A_1+B_1)(A_2+B_2+B_3+F)+A_2(B_2+B_1+F)]_{i'}\left(C_{p,i'} - C_{eq_3,i'}\right)]\}, \qquad i' = \mathrm{H_2 \ or \ CO}$$

where:

$$W_{i'} = \{(A_1+B_1)[A_3(A_2+B_2+B_3+\mathrm{F})+(A_2+B_2)(B_3+\mathrm{F})] + A_2[A_3(B_2+B_3+\mathrm{F})+B_2(B_3+\mathrm{F})]\}_{i'}\,, \quad i' = \mathrm{H_2 \ or \ CO}$$

$$A_{n,i'} = \left[\frac{1}{k_n(1-x_n)^{2/3}}\,\frac{k_{eq,n}}{1+k_{eq,n}}\right]_{i'}, \quad n = 1, 2, 3.$$

$$B_{n,i'} = \left[\frac{(1-x_{n+1})^{1/3}-(1-x_n)^{1/3}}{(1-x_n)^{1/3}(1-x_n)^{1/3}}\,\frac{r_g}{D_{eff,n}}\right]_{i'}, \quad n = 1, 2.$$

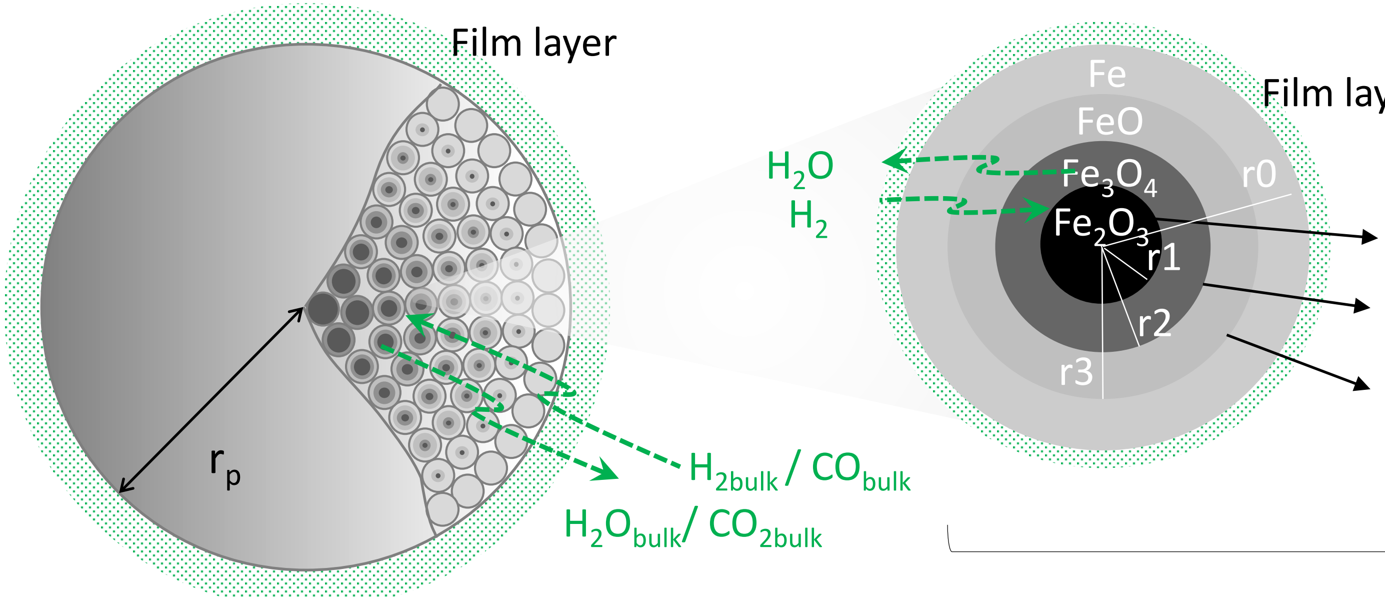
$$B_{3,i'} = \left[\frac{1-(1-x_3)^{1/3}}{(1-x_3)^{1/3}}\,\frac{r_g}{D_{eff,3}}\right]_{i'}$$

$$F_{i'} = \left[\frac{1}{k_g}\right]_{i'}$$

Multiscale Mathematical Modeling

✓ **Pellet model (1D)**

✓ **Grain model (1D)**



Reaction 1: $3\ Fe_2O_3 + H_2/CO \rightleftharpoons 2\ Fe_3O_4 + H_2O/CO_2$

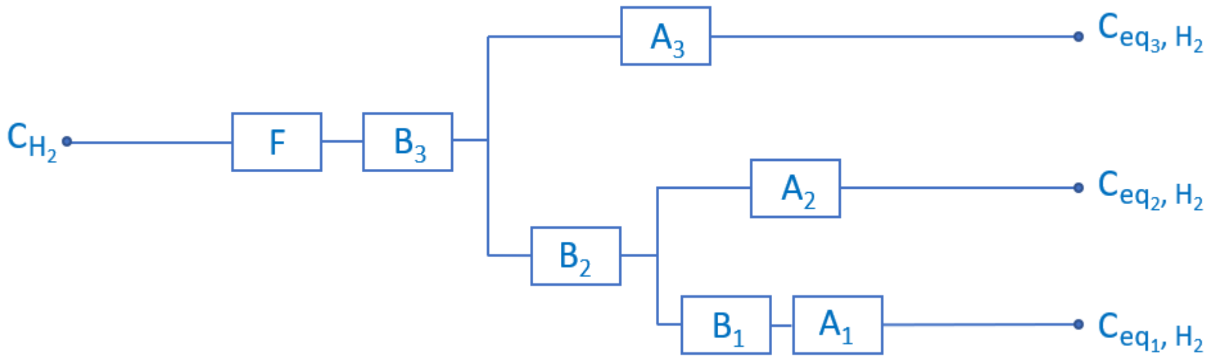
Reaction 2: $Fe_3O_4 + H_2/CO \rightleftharpoons 3\ FeO + H_2O/CO_2$

Reaction 3: $FeO + H_2/CO \rightleftharpoons Fe + H_2O/CO_2$

- Conversion of each solid throughout the pellet : $X_n = \frac{3}{r_p^3}\int_0^{rp} x_n(r,t) r^2 dr$

- Global conversion in the pellet : $X_{global} = \frac{1}{9}X_1 + \frac{2}{9}X_2 + \frac{6}{9}X_3$

- Porosity change: $\varepsilon_p = 1 - N\left(\frac{\rho_{j,0}}{\rho_{j,f}}\right)(1 - \varepsilon_{j,0})$



Main equation for multi-scale modeling

✓ Moving-bed reactor scale model (2D)

- Mass balance for gas (i= H_2, H_2O, CO, CO_2, N_2)

$$-D_i\nabla^2C_i+\nabla(C_iu_g)=-(1-\varepsilon_b)A_pD_{i,eff}\frac{dC_{i,p}}{dr}|_{r=r_p}$$

- Mass balance for solid (j= Fe_2O_3, Fe)

$$\frac{dC_j}{dz}=\frac{3}{{r_p}^3}\int_0^{r_p}C_{j,p}\,r^2dr\,(1-\varepsilon_b)$$

- Heat balance for gas

$$-k_g\nabla^2T_g+\rho_gCp_g\nabla(u_gT_g)=-(1-\varepsilon_b)A_ph\left(T_g-T_s|_{r=r_p}\right)$$

- Brinkman equation

$$\frac{\rho}{\varepsilon_b}\left((u_g.\nabla)\frac{u_g}{\varepsilon_b}\right)$$
$$=-\nabla P+\nabla.\left\{\frac{1}{\varepsilon_b}\left[\mu\left(\nabla u_g+(\nabla u_g)^T\right)-\frac{2}{3}\mu(\nabla.u_g)I\right]\right\}-\left(\kappa^{-1}\mu+\frac{Q_m}{{\varepsilon_b}^2}\right)u_g+F$$

✓ Pellet scale model (1D)

- Mass balance for gas (i= H_2, H_2O, CO, CO_2, N_2)

$$-D_{i,eff}\frac{\partial^2C_i}{\partial r}=\frac{2}{r}D_{i,eff}\frac{dC_{p,i}}{dr}+R_i(r,z)$$

- Mass balance for solid (j= Fe_2O_3, Fe)

$$\nabla(C_{p,j}u_s)=(1-\varepsilon_b)R_i(r,z)$$

- Heat balance for solid

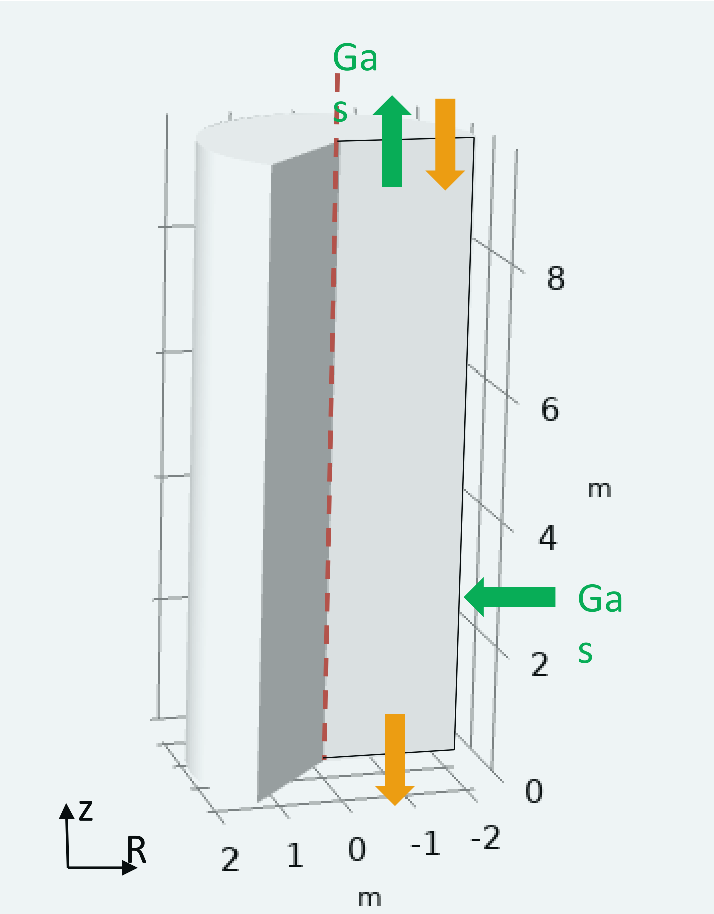
$$-k_{ceff,p}\nabla^2T_s+\rho_pCp_{eff,p}\nabla(u_sT_{gs})-\frac{1}{r^2}\frac{d}{dr}\left(r^2k_{ceff,p}\frac{dT_s}{dr}\right)$$
$$=\sum_{j=1}^{NR}R_j(-\Delta H_n)$$

$$\left.\begin{matrix}k_{ceff,p}\\ Cp_{eff,p}\end{matrix}\right\}$$ Solid + gas in the pellet

Multiscale Mathematical Modeling

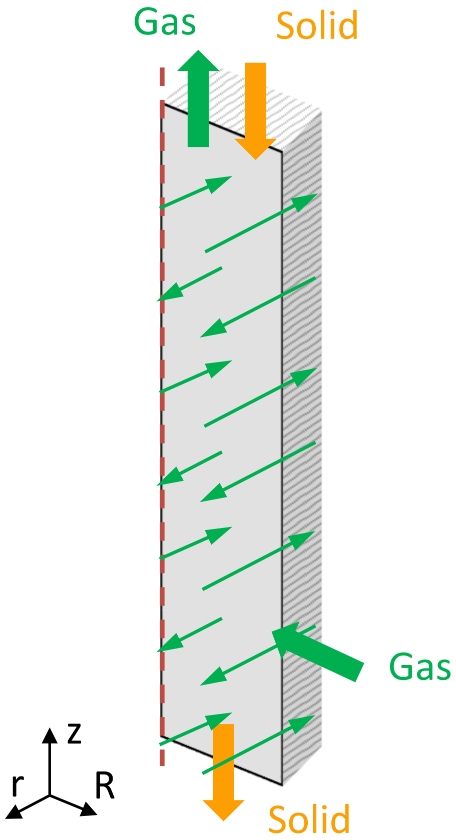
Reactor Scale

2D-axissymmetric geometic



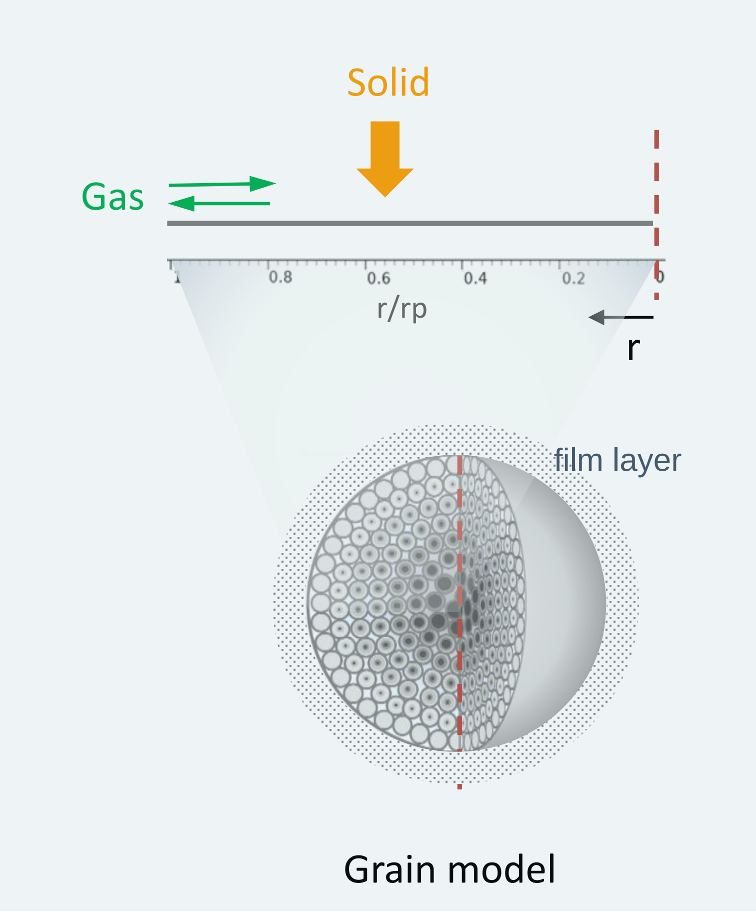
Reactor-pellet scale coupling

Extra dimension



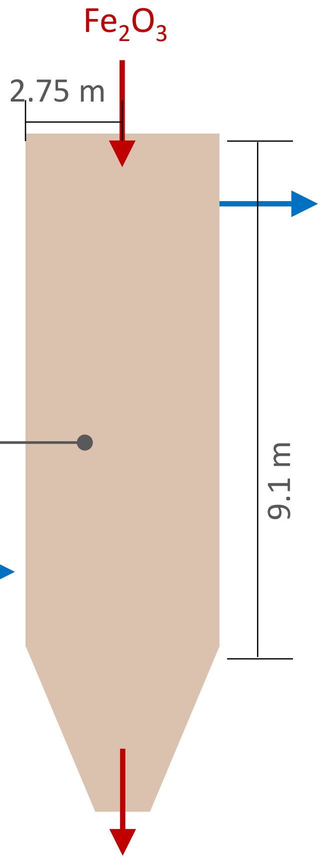
Pellet Scale

1D symmetric geometric



Industrial Plant Data

Morabake MIDREX Plant (Nouri et al, 2011)



Entrada de sólidos

Densidade da pelota	4.7	g/cm³
Diâmetro da pelota	11	mm
Temperatura	300	K
Porosidade da pelota	0.15	

Propriedades do leito

Pelotas/volume	0.64	pellet/cm³
Densidade do leito	2	g/cm³

Entrada de gases

Vazão molar	177180	Nm³/h
Pressão	1.35	bar
Temperatura	1174	K

Composição dos gases de entrada		
H_2	53.47	%
CO	34.45	%
H_2O	5.83	%
CO_2	2.6	%
CH_4+N_2	3.65	%

Gases de saída

Pressão	131325	Pa
Temperatura	791	K

Composição dos gases de saída		
H_2	32.24	%
CO	21.6	%
H_2O	25.05	%
CO_2	15.46	%
CH_4+N_2	3.65	%

Saída dos sólidos

Metalização	94.8	%
Produção de Fe	110	t/h

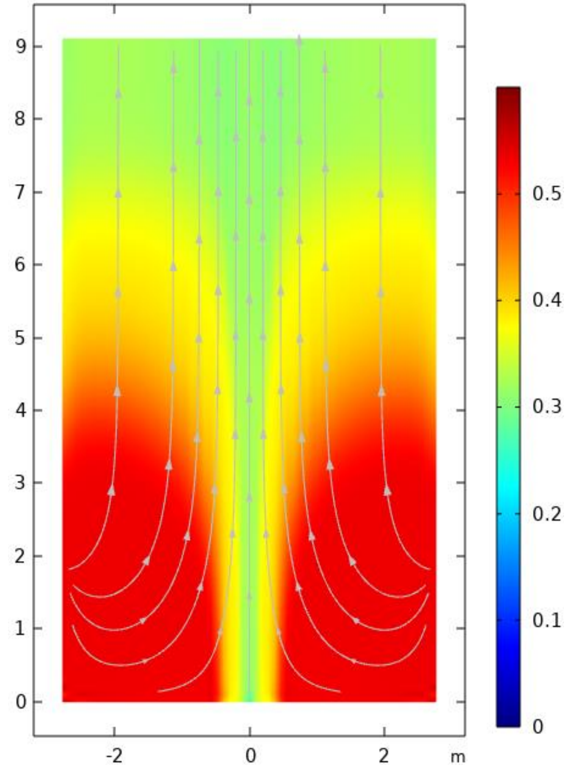
Results and Discussion

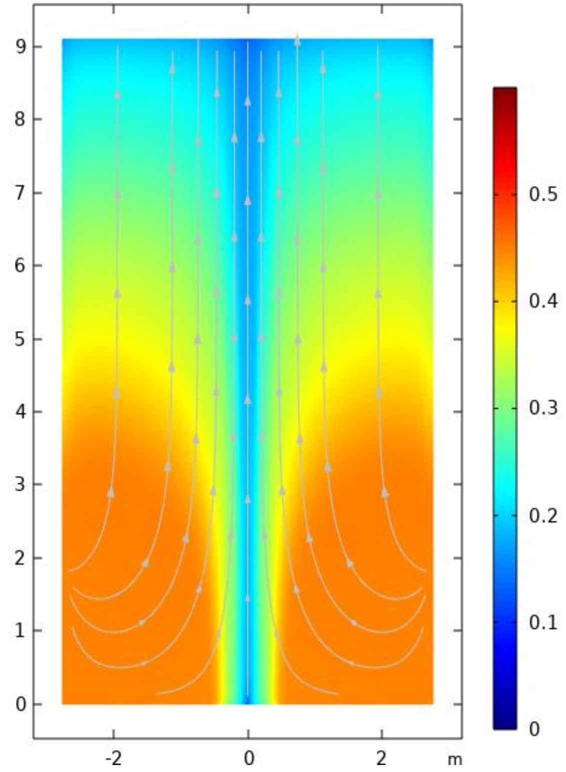
Comparison between the data from Morabake Plant and model

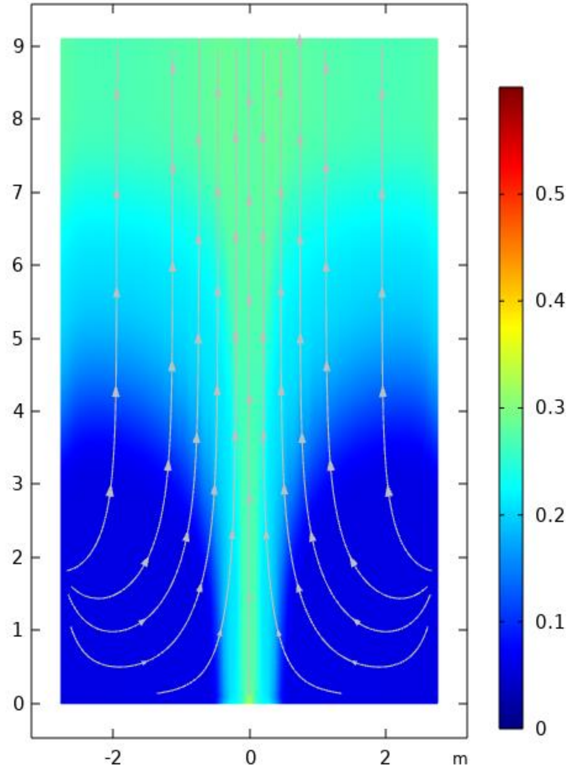
Measured parameters	**Plant data**	**Model data results**	**Error**
Outlet gas composition			
H_2	32.24 %	32.11 %	0.40 %
CO	21.6 %	21.59 %	0.05 %
H_2O	25.05 %	27.2 %	8.58 %
CO_2	15.46 %	15.45 %	0.06 %
$CH_4 + N_2$	3.65 %	3.65 %	0 %
Temperature	791 K	760 K	3.92 %
Solid conversion	94.8 %	96.5 %	1.79 %

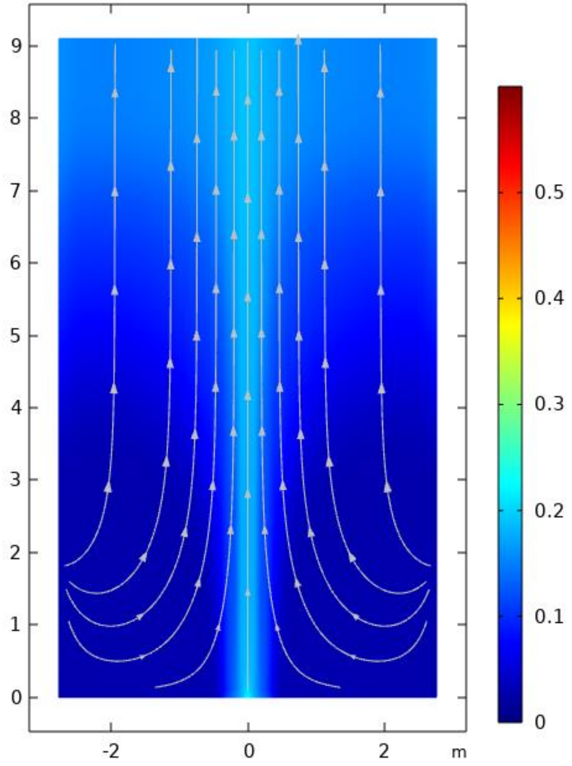
Results and Discussion

Gas mole fraction distribution at reactor scale


H_2 mole fraction


CO mole fraction


CO mole fraction


CO_2 mole fraction

Results and Discussion

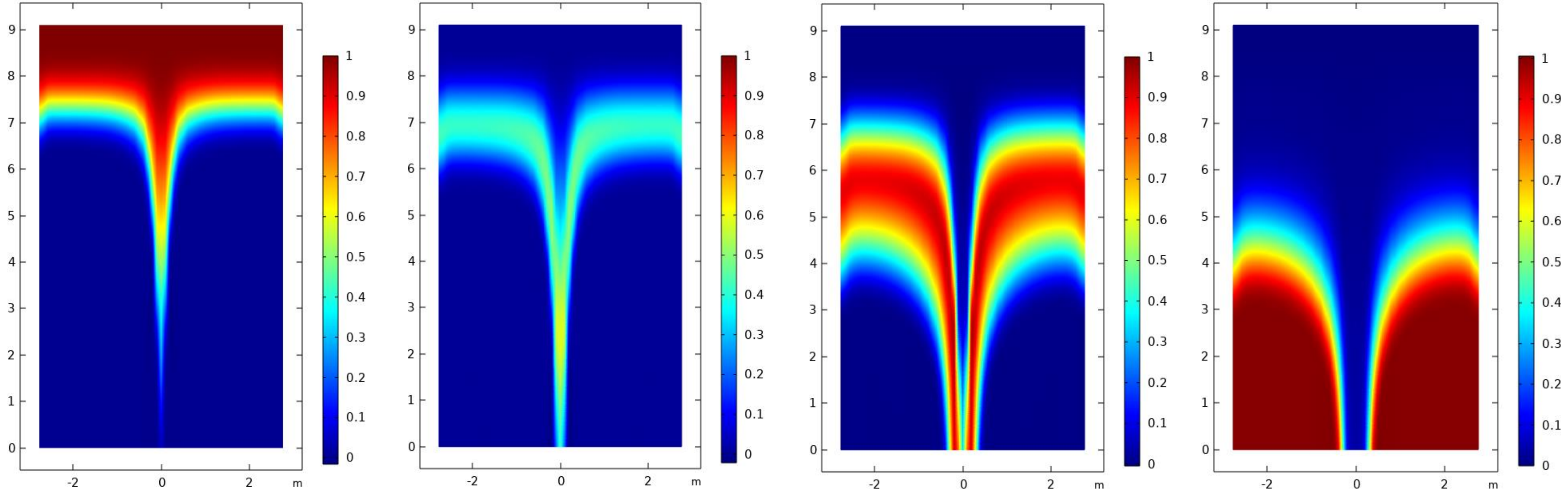
Solid mole fraction distribution at reactor scale

Fe_2O_3 mole fraction

Fe_3O_4 mole fraction

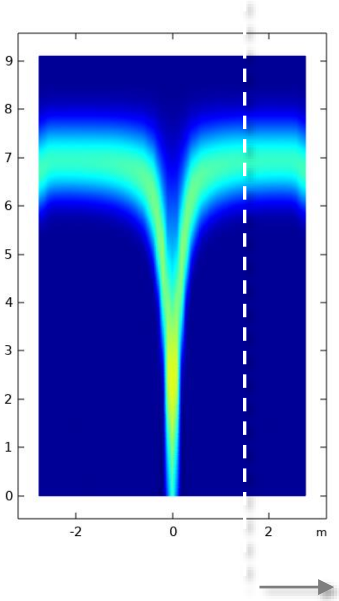
FeO mole fraction

Fe mole fraction

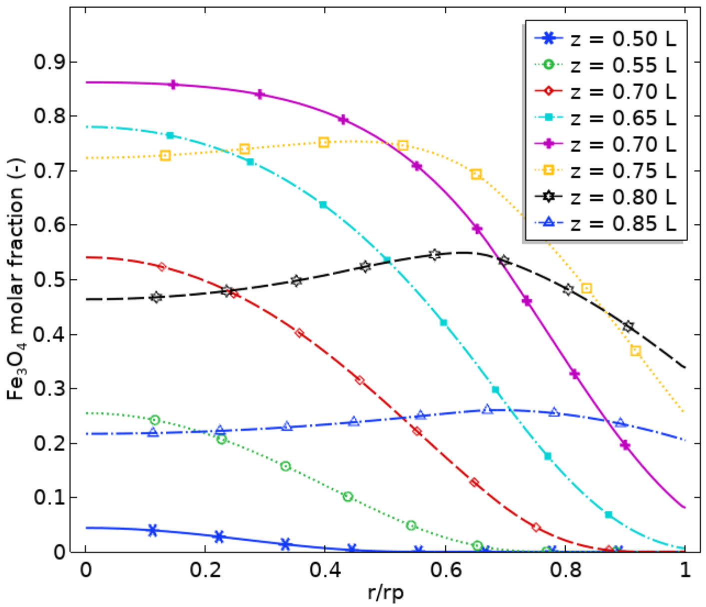


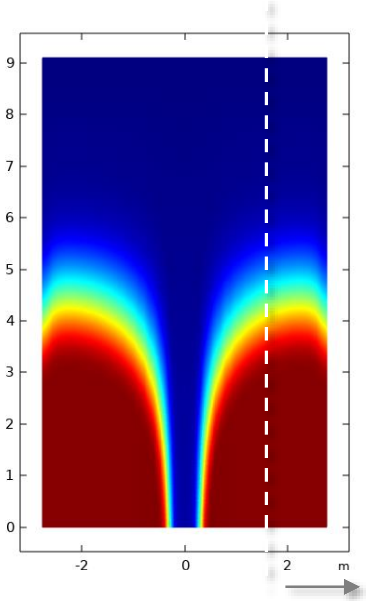
Results and Discussion

Solid mole fraction distribution at pellet scale

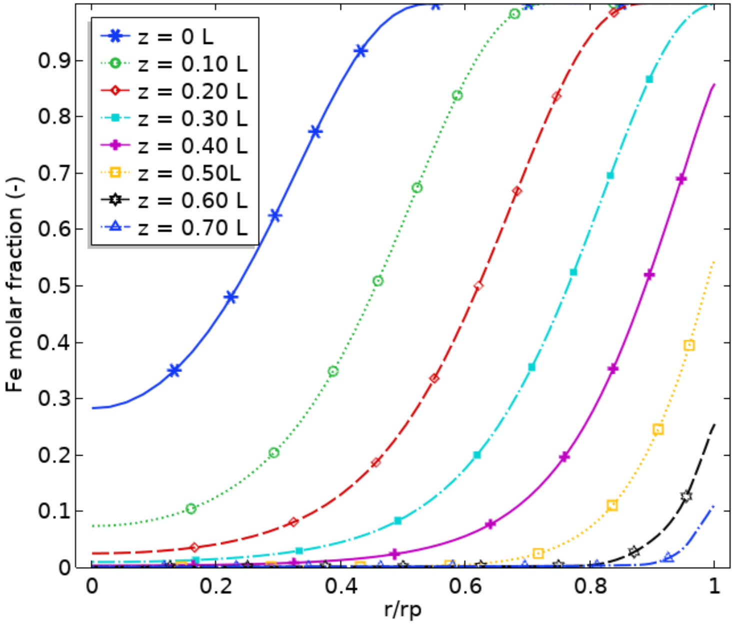


Fe_3O_4 mole fraction





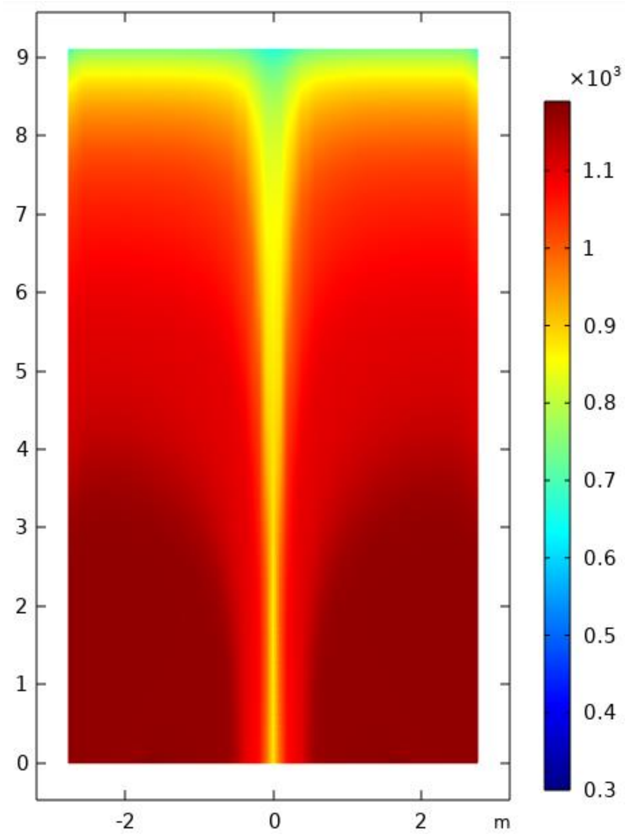
Fe mole fraction



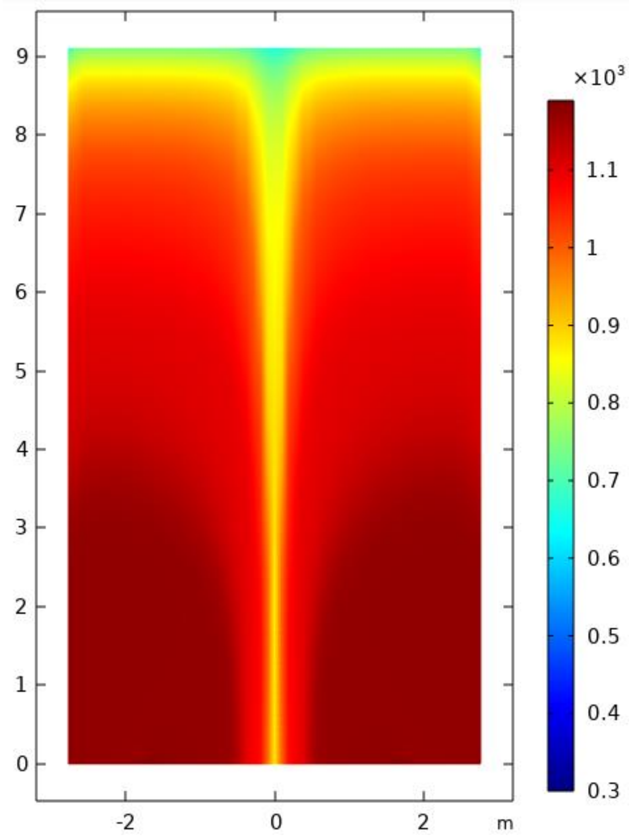
Results and Discussion

Temperature distribution at reactor

Gas temperature (K)



Solid temperature (K)



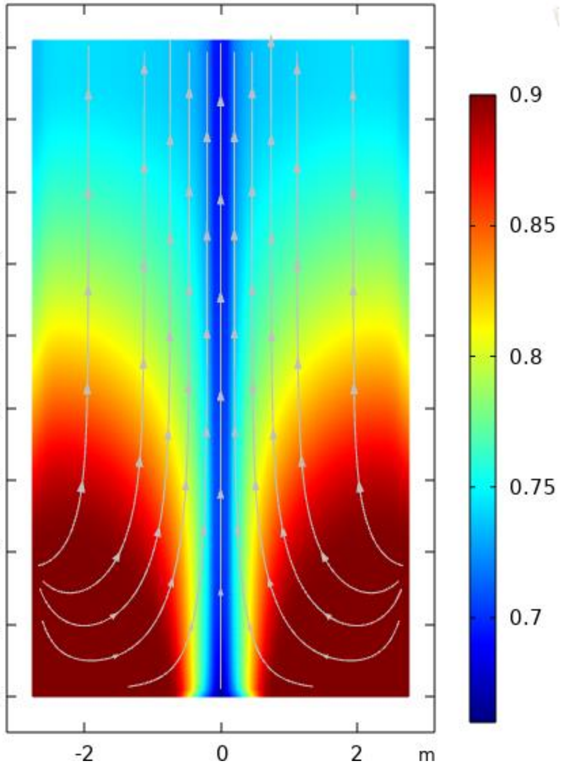
Temperature distribution at pellet scale

Isothermal pellet

Biot number < 0.1

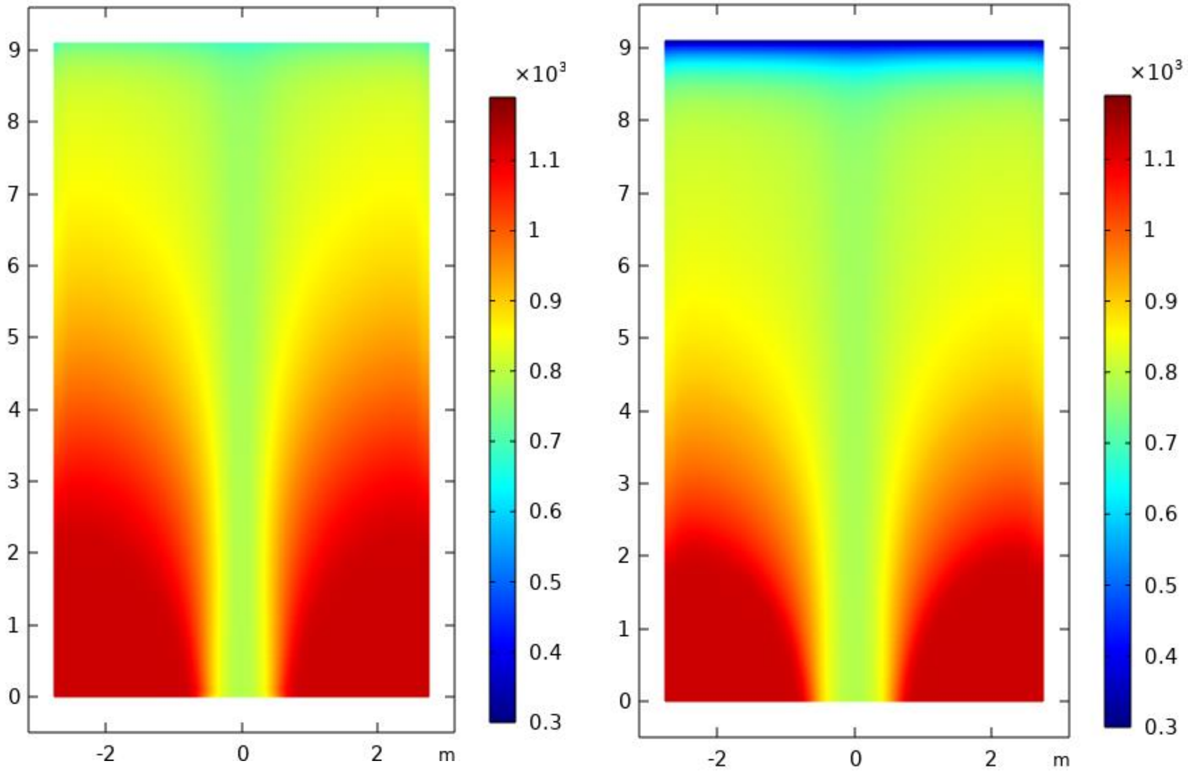
Predictions for Hydrogen Direct Reduction process

Gas mole fraction distribution at reactor scale


H_2 mole fraction

H_2O mole fraction

Temperature at reactor scale

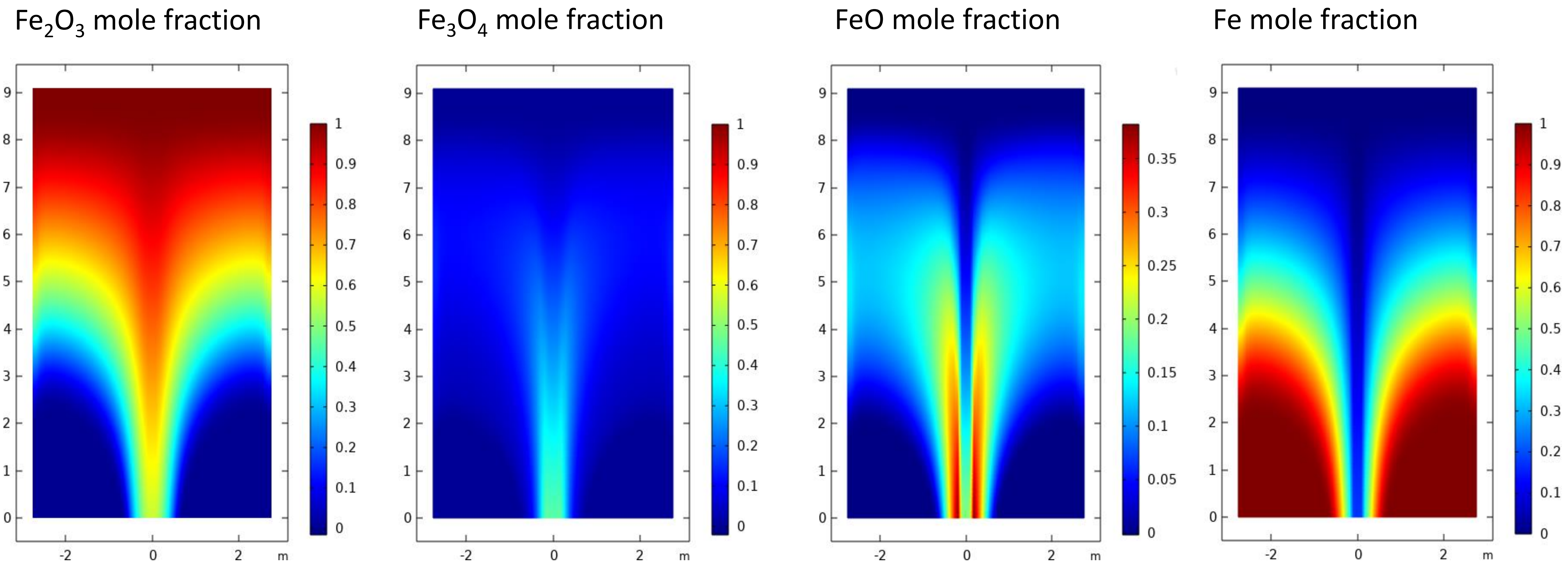

Gas temperature (K)

Solid temperature (K)

4 x the stoichiometric value required for the global reduction from Fe_2O_3 to Fe (3 moles H_2/1 mole Fe_2O_3)

Results and Discussion

Solid mole fraction distribution at reactor scale



Conclusions

- ✓ Multiscale model presents a good predictive capacity for the industrial direct reduction process.
- ✓ non-homogeneous gas flow field inside the reactor.
- ✓ Improve the partial reduction of the solid phase at the reactor center.
- ✓ Predictions for Hydrogen direct reduction.

THANK YOU !

Institute for Tecnhological Research (IPT)

Escola Politécnica da Universidade de São Paulo (Poli - USP)

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