MODELING ISSUES FOR THE NUMERICAL SIMULATION OF THE HYDRODYNAMIC FOR THE CHEMICAL LOOPING

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Research objectives and strategy

- Explore and model local interactions and medium scale behavior in reactive and/or multiphase flows with dispersed phases of solid particles or droplets by using experiments and direct numerical simulations.

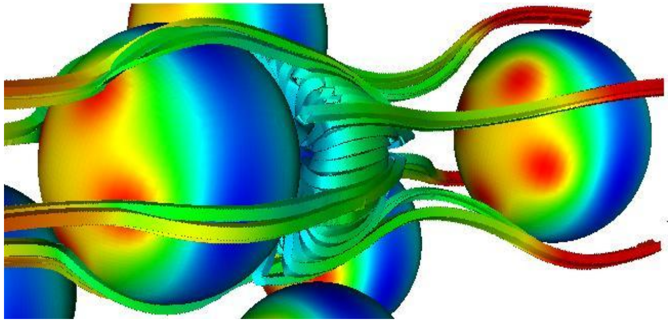
- Develop numerical modeling approaches for full-scale predictions of reactive particulate multiphase flows in the general frame of kinetic theory of particulate flows:
  - fluid-particle joint probability density function (PDF) equation,
  - "n-fluid" (or moment) and stochastic Lagrangian (or Monte Carlo) methods coupled with RANS fluid equations,
  - Euler-Euler and Euler-Lagrange large-eddy simulation (LES) approaches

- Full scale prediction of industrial (and environmental) flows:
  - Evaluation of available numerical modeling approaches (comparison with experimental results),
  - Optimization and scale-up of existing processes,
  - Support for development of new processes.

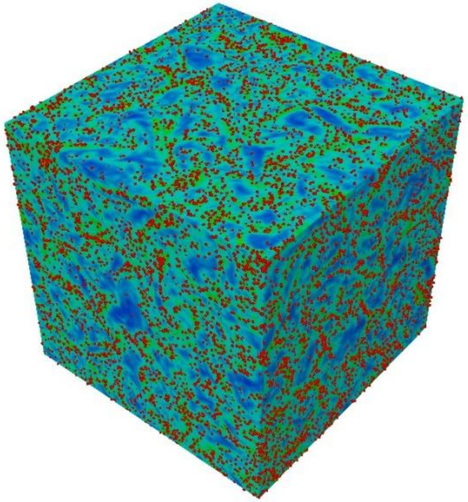
Research objectives and strategy

Microscopic scale (~1*mm*)
DNS, immersed boundary method

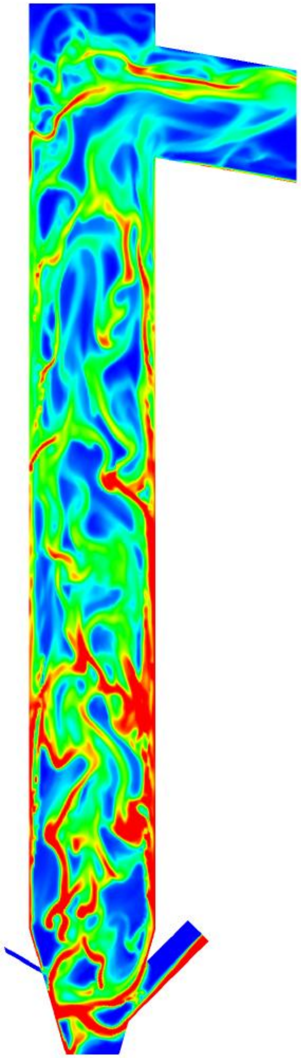
Movie



Mesoscopic scale (~10*cm*)
DNS/LES + Discrete Particle Simulation (DPS)



Macroscopic scale (~10*m*)
RANS, Euler-Euler



MeOx concentration field. NEPTUNE_CFD bi-solid prediction of the circulating fluidized bed coal combution reactor (chemical looping fuel reactor)

Eulerian approach for dense particulate flows

Kinetic theory of polydispersed solid particle mixture:

Closure of the kinetic transport equation on the single particle PDF based on a Lagrangian modeling of particle-fluid, particle-particle and particle-wall interactions.

$$\frac{\partial f_{fp}}{\partial t}+\frac{\partial}{\partial x_i}\left[c_{p,i}f_{fp}\right]+\frac{\partial}{\partial c_{p,i}}\left[\left\langle\frac{du_{p,i}}{dt}|\mathbf{c}_f,\mathbf{c}_p\right\rangle f_{fp}\right]+\frac{\partial}{\partial c_{f,i}}\left[\left\langle\frac{du_{f@p,i}}{dt}|\mathbf{c}_f,\mathbf{c}_p\right\rangle f_{fp}\right]=\left(\frac{\partial f_{fp}}{\partial t}\right)_{coll}$$

Derivation of the moment transport equations (concentration, velocity, temperature, fluctuating motion kinetic energy, kinetic stresses…) and the transport properties (viscosity, diffusivity).

Validation from Euler-Lagrange “numerical experiments“

→ *Implementation in NEPTUNE_CFD and comparison of model predictions with experimental measurements (laboratory, pilot and industrial scales).*

Mathematical model

Multi-fluid model, implemented in NEPTUNE CFDV108@Tlse, developed in the frame of kinetic approach with additional equation accounting for the effect of the interstitial fluid.

- Mass balance equation

$$\frac{\partial}{\partial t}(\alpha_k \rho_k) + \frac{\partial}{\partial x_j}(\alpha_k \rho_k U_{k,j}) = 0$$

+ source terms for chemical looping

- Momentum balance equation

$$\alpha_k \rho_k \left[\frac{\partial U_{k,i}}{\partial t} + U_{k,j}\frac{\partial U_{k,i}}{\partial x_j}\right] = -\alpha_k \frac{\partial P_g}{\partial x_i} + \alpha_k \rho_k g_i + \sum_{q=g,p} I_{q\to k,i} - \frac{\partial \Sigma_{k,ij}}{\partial x_j}$$

+ source terms for chemical looping

Gas-particle momentum transfer (Gobin et al, 2003)

$$I_{g\to p,i} = -I_{p\to g,i} = -\alpha_p \rho_p \frac{V_{r,i}}{\tau^F_{gp}}$$

$$\frac{1}{\tau^F_{gp}} = \frac{3}{4}\frac{\rho_g}{\rho_p}\frac{\langle|\mathbf{v}_r|\rangle}{d_p}\min(C_{D,WY}, C_{D,Erg})$$

particle relaxation time

$$Re_p = \frac{\alpha_g d_p \langle|\mathbf{v}_r|\rangle}{\nu_g}$$

particle Reynolds number

$$V_{r,i} = U_{p,i} - U_{g,i} - V_{d,i}$$

mean gas-particle relative velocity

Particle-particle momentum transfer:

$$I_{q\to p,i} = -\frac{m_p m_q}{m_p + m_q}\frac{1+e_c}{2}\frac{n_p}{\tau^c_{pq}} H_1(z)\left(U_{p,i} - U_{q,i}\right)$$

Mathematical model

- <u>Turbulence modeling:</u>
  - *LES* model for the gas
  - Two-equations model for the particles (q_p^2-q_{gp})

- <u>Effective solid stress modeling</u>

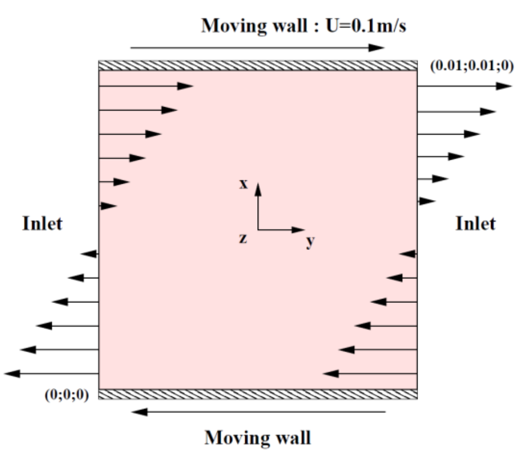
$$\Sigma_{p,ij} = \left[P_p - \lambda_p \frac{\partial U_{p,m}}{\partial x_m}\right]\delta_{ij} - \mu_p \left[\frac{\partial U_{p,i}}{\partial x_j} + \frac{\partial U_{p,j}}{\partial x_i} - \frac{2}{3}\frac{\partial U_{p,m}}{\partial x_m}\delta_{ij}\right]$$

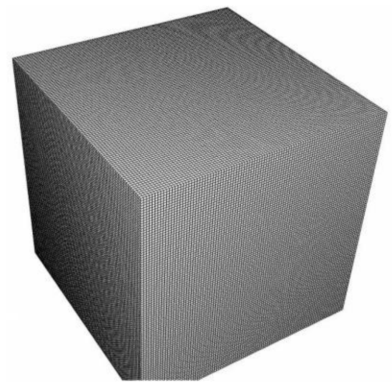
$$\mu_p = \alpha_p \rho_p (\nu_p^{kin} + \nu_p^{col}) \quad \left\{ \begin{array}{l} \nu_p^{kin} = \left[\frac{1}{3} q_{gp}\tau_{gp} + \frac{1}{2}\tau_{gp}^F \frac{2}{3} q_p^2 \left(1 + \hat{\alpha}_p g_0 \Phi_c\right)\right] \times \left[1 + \frac{\tau_{gp}^F}{2}\frac{\sigma_c}{\hat{\tau}_p^c}\right]^{-1} \\ \nu_p^{col} = \frac{4}{5}\hat{\alpha}_p g_0 (1 + e_c)\left[\nu_p^{kin} + \hat{d}_p \sqrt{\frac{2}{3}\frac{q_p^2}{\pi}}\right] \end{array} \right.$$

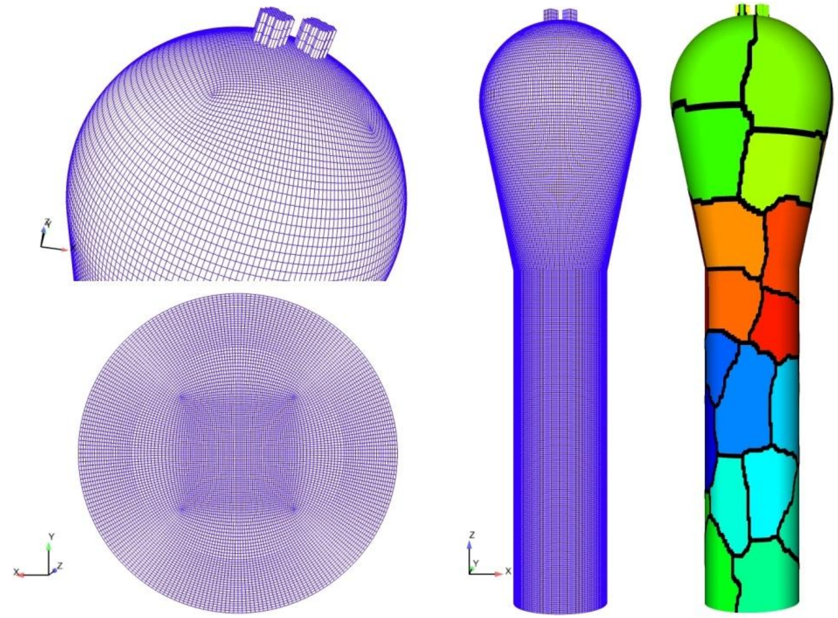
The polydispersion (Batrak et al., 2005)

$$\hat{\alpha}_p = \sum_{q \neq p} \alpha_p \frac{2 m_q}{m_p + m_q}\left[\frac{d_{pq}}{d_q}\right]^3 \qquad \hat{d}_p = \frac{1}{\hat{\alpha}_p}\sum_{q \neq p} \alpha_q \frac{d_{pq}^4}{d_q^3}\frac{2 m_q}{m_p + m_q} \qquad \frac{1}{\hat{\tau}_p^c} = \sum_{q \neq p} \frac{2 m_q}{m_p + m_q}\frac{1}{\tau_{pq}^c}$$

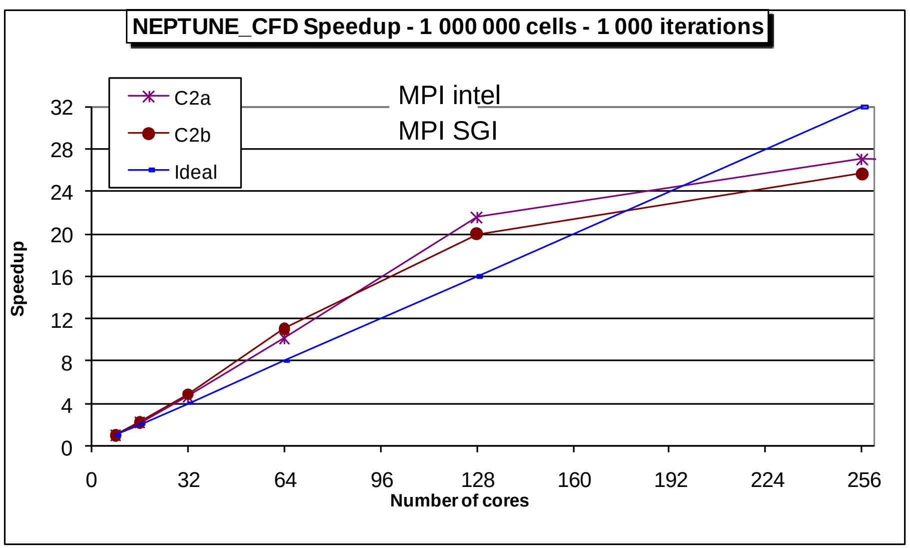
NEPTUNE_CFD computation efficiency:

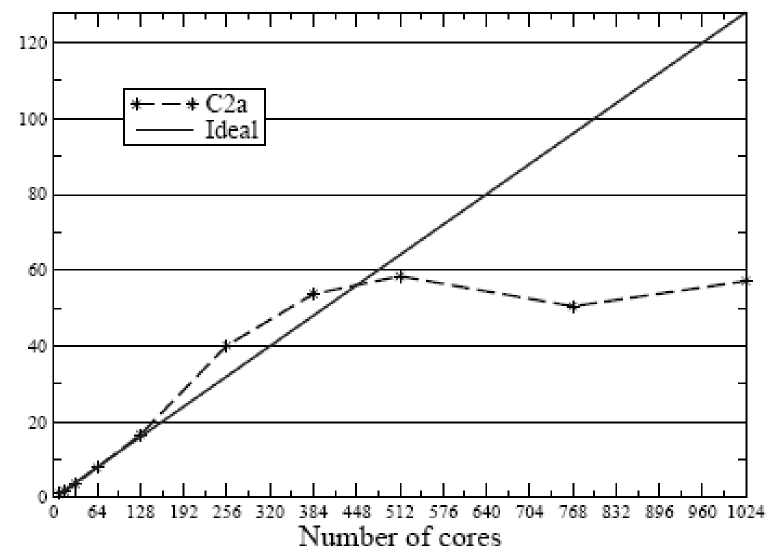




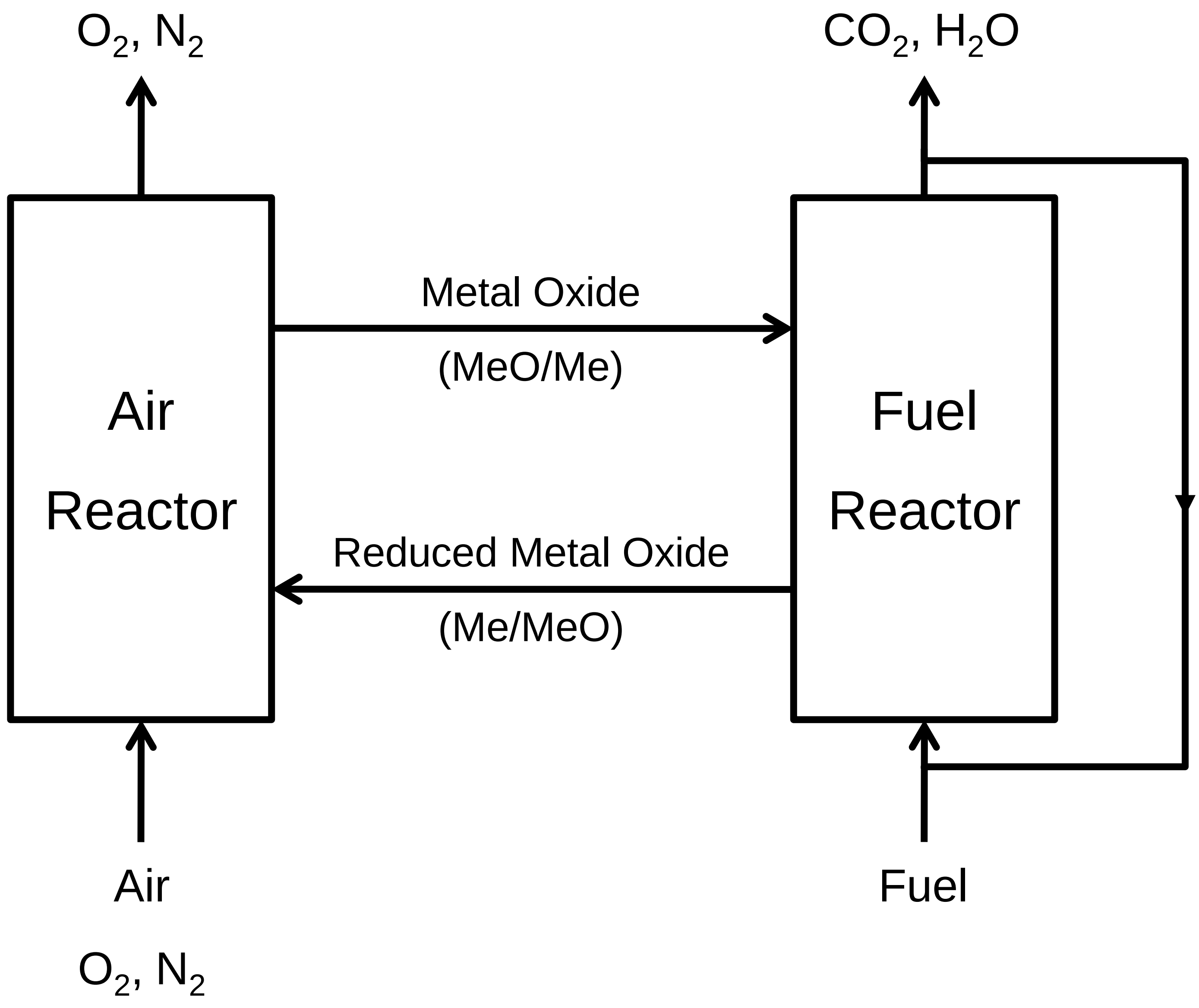


3,150,716 cells





Chemical looping



Chemical looping

Utilization of 3D CFD in the frame of industrial project:

Computation of an experimental cold gas-solid circulating fluidized bed (UTC) and comparison with experimental data (pressure drop):

- → influence of the operating conditions (solid and gas fluidization velocity),
- → influence of the amount of fines on the recirculation of coarse particles,
- → influence of secondary air injection and comparison with volumetric source term representing the particle gasification.

Evaluation of ash/MeOx separation in dense bubbling fluidized bed

- → validation of the concept (industrial scale)
- → correlation derivation for solid separation effect (laboratory and industrial scales)

Numerical simulation of medium-scale pilot (1MW)

Chemical looping

Peculiar fluidized bed operating conditions:

Mixture of particle species with a large contrast in diameter and density:

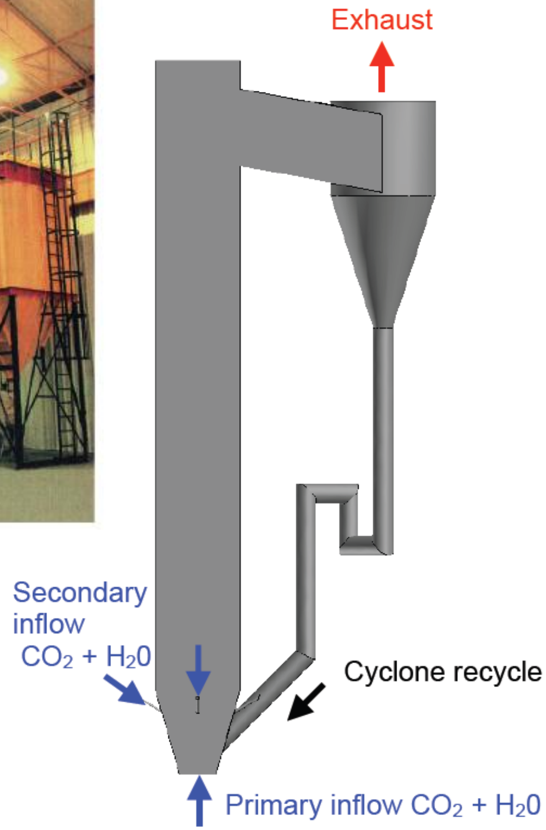
- MeOx particles: $d_p \sim 200$ mm, $\rho_p \sim 5\times10^3$ kg/m^3
- Coal (+ ash) particles: $d_p < 50$ mm, $\rho_p \sim 1\times10^3$ kg/m^3

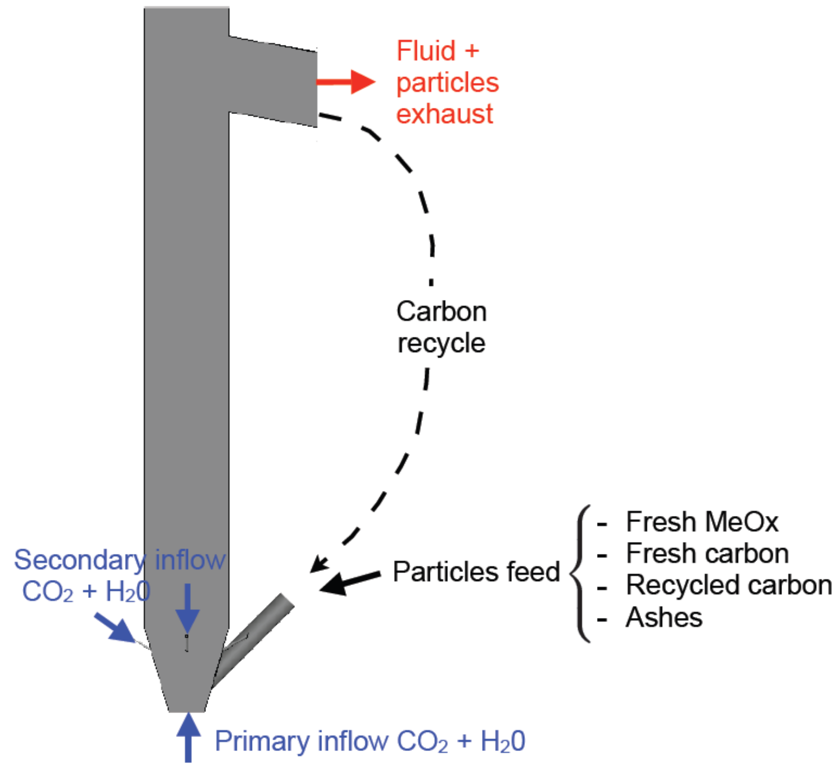
→ Particle relaxation time ratio: > 50

→ Particle settling velocity ratio: > 40

Local effective production of gas in the fuel reactor.

Prediction of solid mass flux



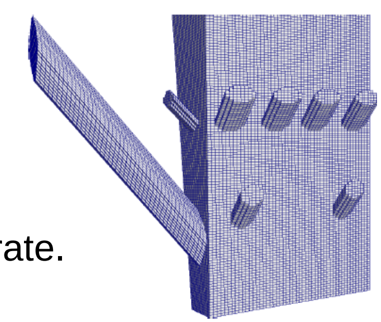


Given conditions:

- Gas fluidization and fresh carbon injection,
- MeOx solid inventory,
- Carbon reaction rate,
- Cyclone « cut-off » diameter.

Results:

- Pressure drop,
- MeOx circulating mass flow rate.



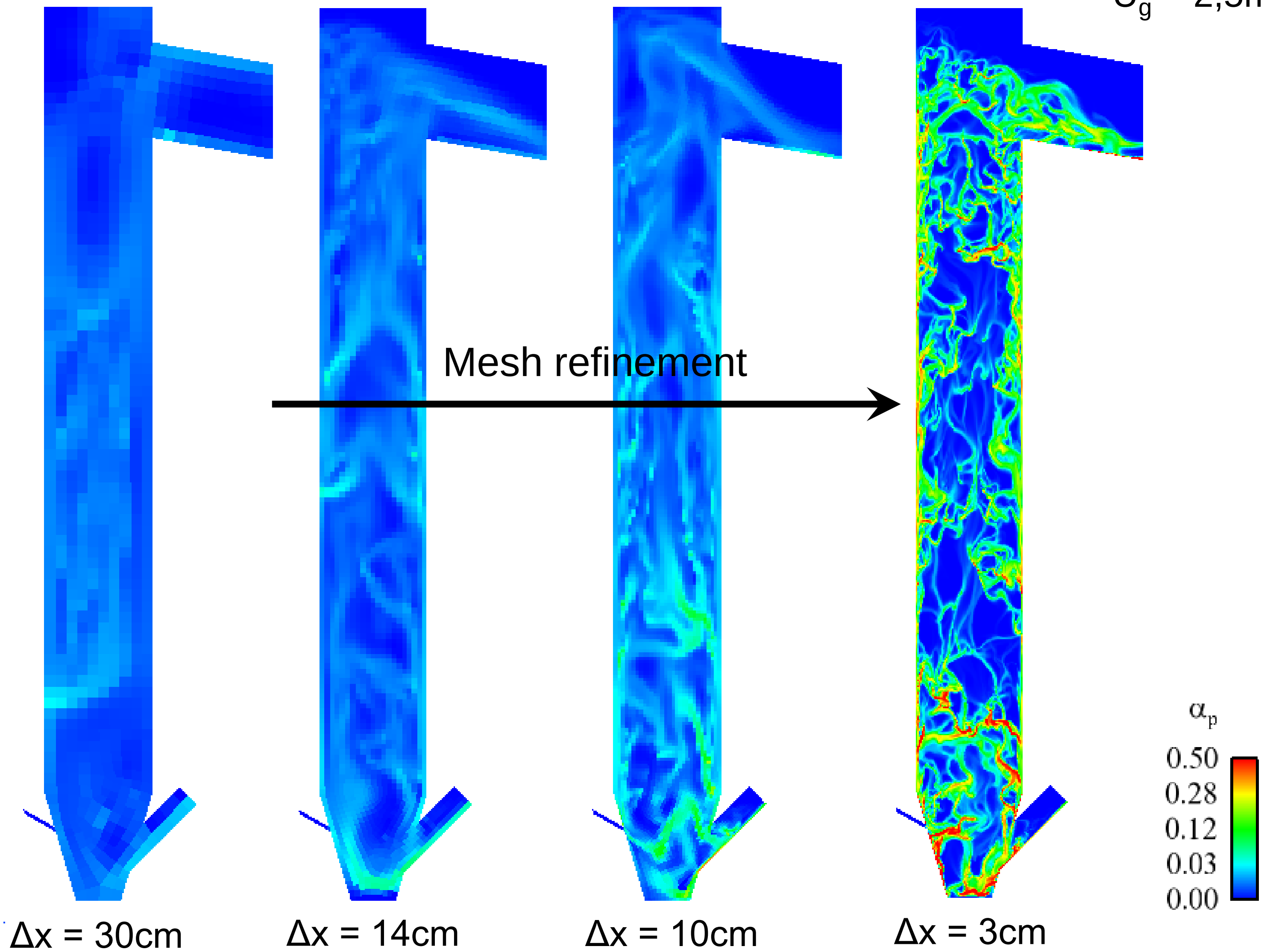
Prediction of solid mass flux

Influence of the mesh refinement:

$d_p \sim 140\mu m$

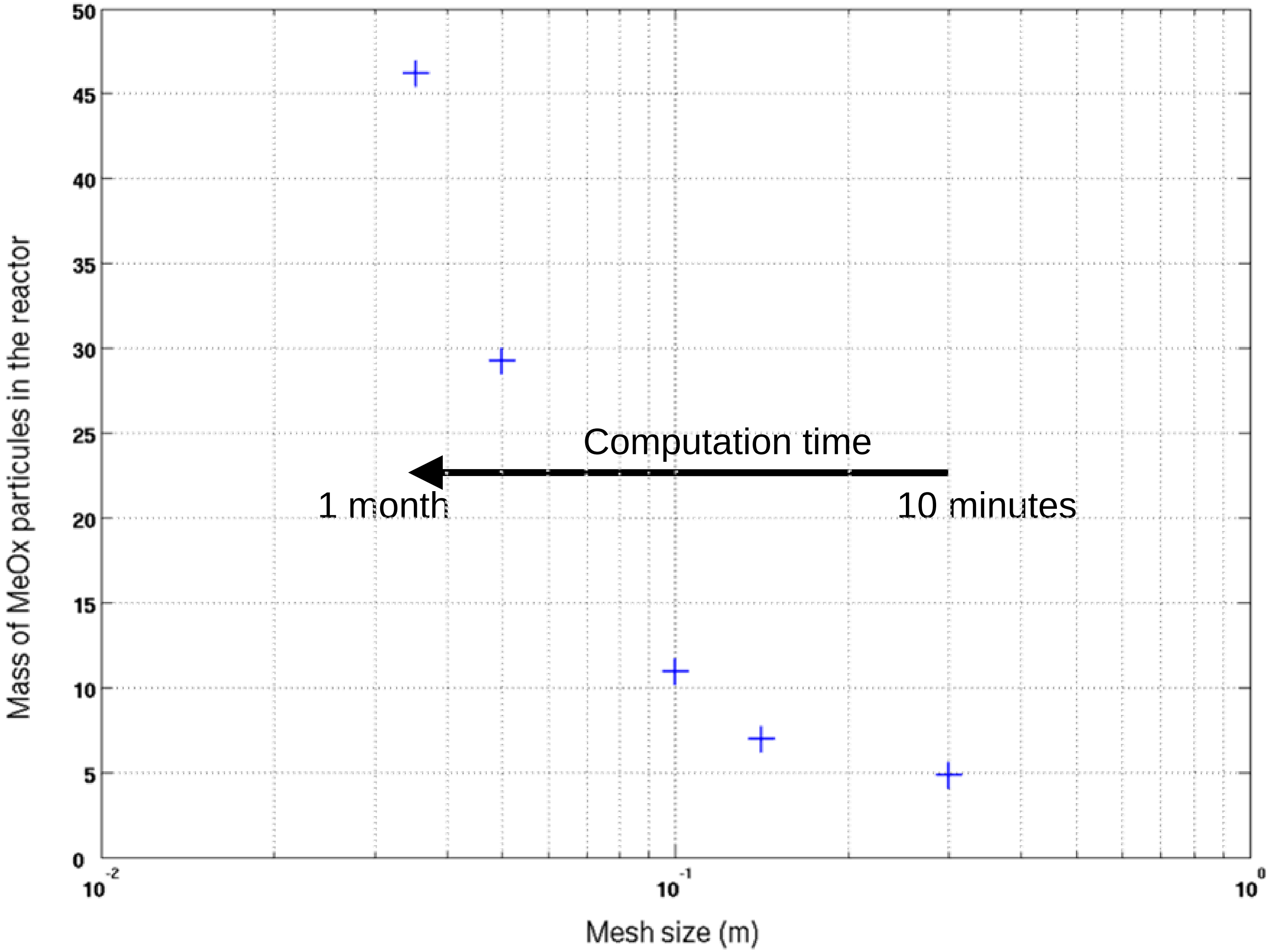
$\rho_p/\rho_g \sim 11000$

$U_g \sim 2{,}5 m/s$



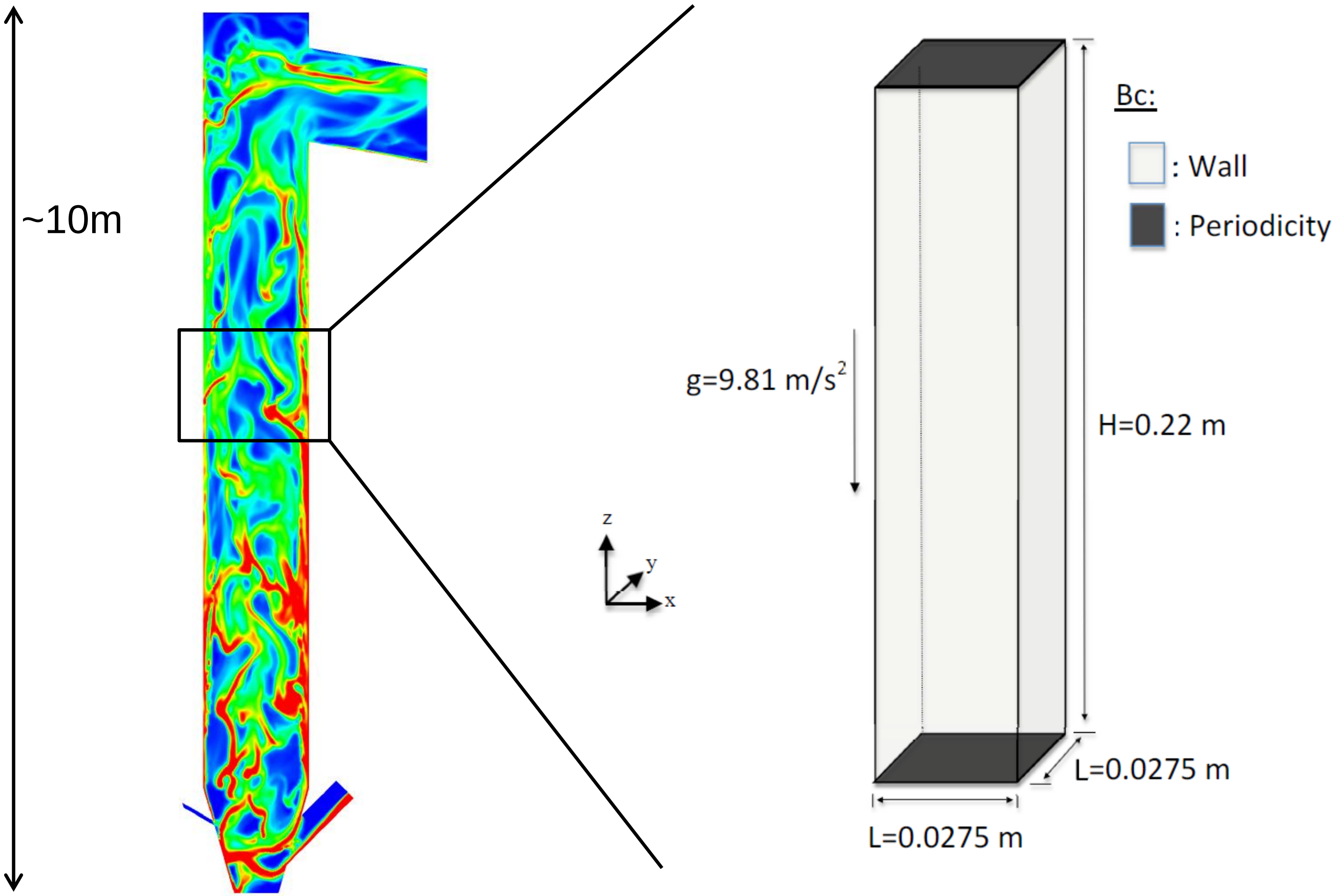
Prediction of solid mass flux

Influence of the mesh refinement:



Prediction of solid mass flux

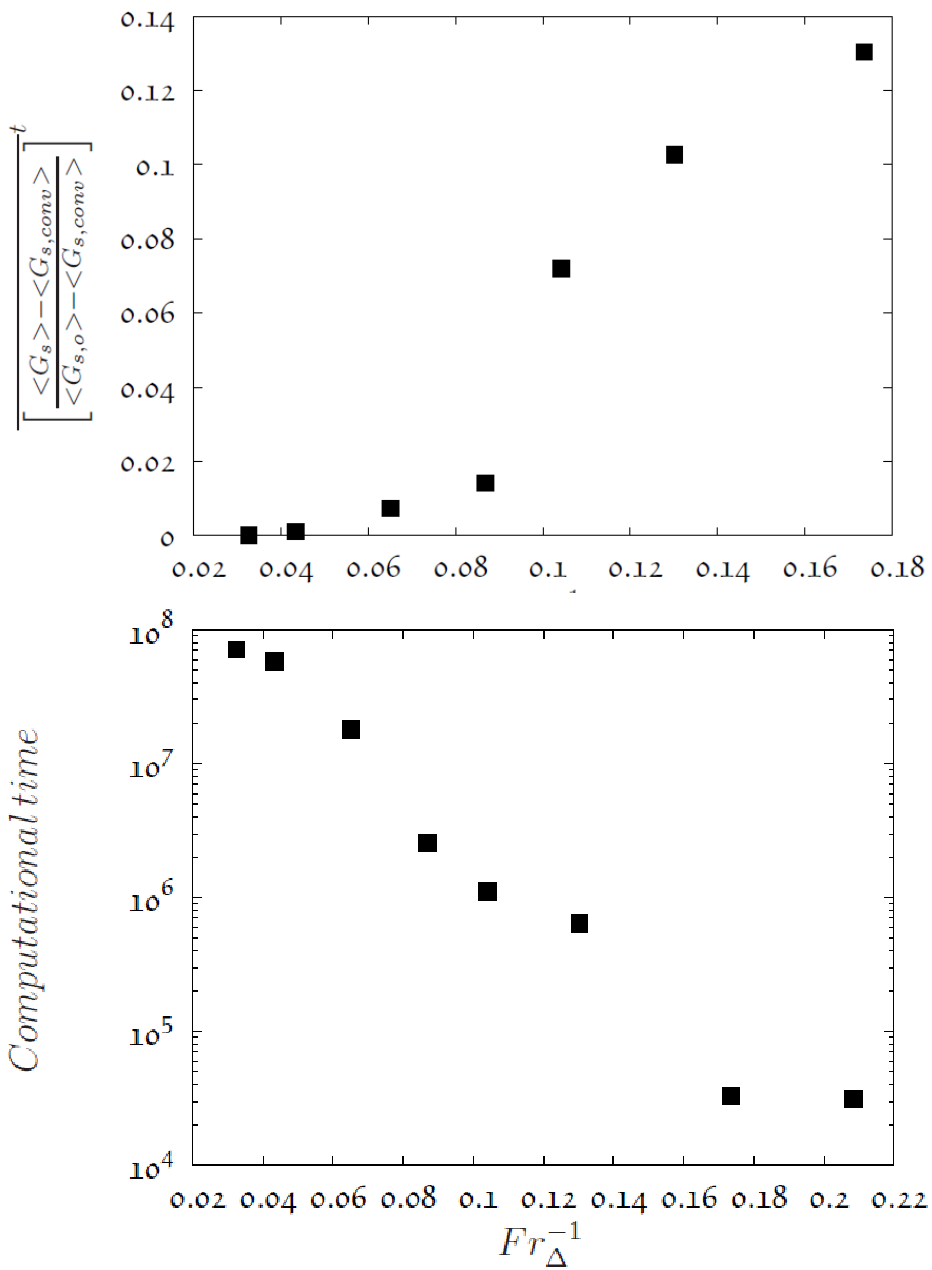
Mesh independent simulation:



Prediction of solid mass flux

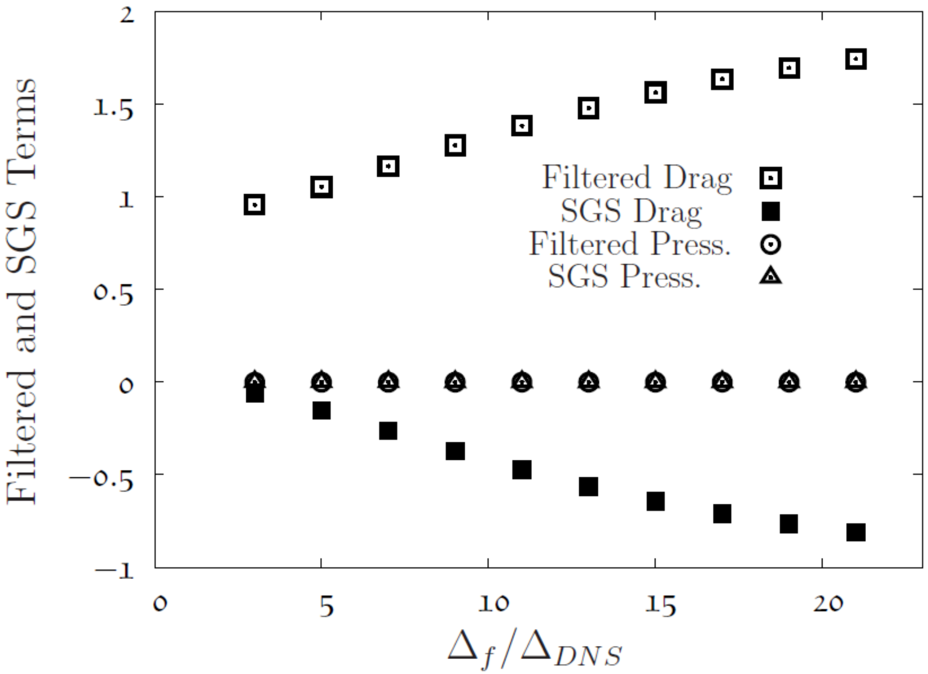
Mesh independent simulation:

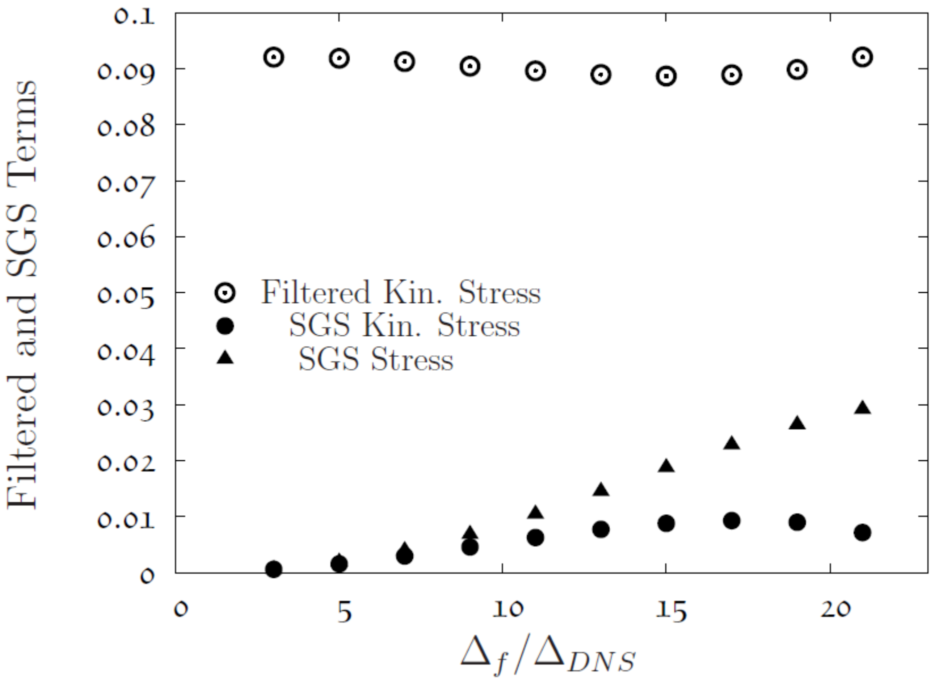
Mesh-converged 3d numerical simulations with NEPTUNE_CFD (18 000 000 cells)



Prediction of solid mass flux

Budget analysis:





Prediction of solid mass flux

Subgrid model development ("Two-fluid LES approach"):

Parmentier et al.,AIChE J., 2011

Filtered drag term

$$\overline{\frac{\alpha_p\rho_p\left(U_{p,i}-U_{p,i}\right)}{\tau_{gp}^F}} \approx \frac{\overline{\alpha}_p\rho_p}{\overline{\tau}_{gp}^F}\left(\widetilde{U}_{p,i}-\widetilde{U}_{g,i}+\widetilde{V}_{d,i}\right)$$

Subgrid velocity: $\widetilde{V}_{d,i}=\widetilde{U}_{g,i}-\widetilde{U}_{g@p,i}$

Subgrid drift velocity model

General form: $\widetilde{V}_{d,i}=g_{ij}(\overline{\alpha}_p,\widetilde{Re}_p,\Delta/\Delta_c)\times\left(\widetilde{U}_{p,j}-\widetilde{U}_{p,j}\right)$

with $\widetilde{Re}_p=\frac{\overline{\alpha}_g d_p|\widetilde{V}_r|}{\mu_g}$, Δ the filter width and Δ_c the cell width

Model #1:

$$\widetilde{V}_{d,i}=g(\overline{\alpha}_p\widetilde{Re}_p,\Delta/\Delta_c)\times\widetilde{V}_{r,i}$$

Model #2:

$$\overline{\alpha}_p\widetilde{V}_{d,i}=K_{ij}\ f(\Delta/\Delta_c)h(\overline{\alpha}_p)\widetilde{V}_{r,j}$$

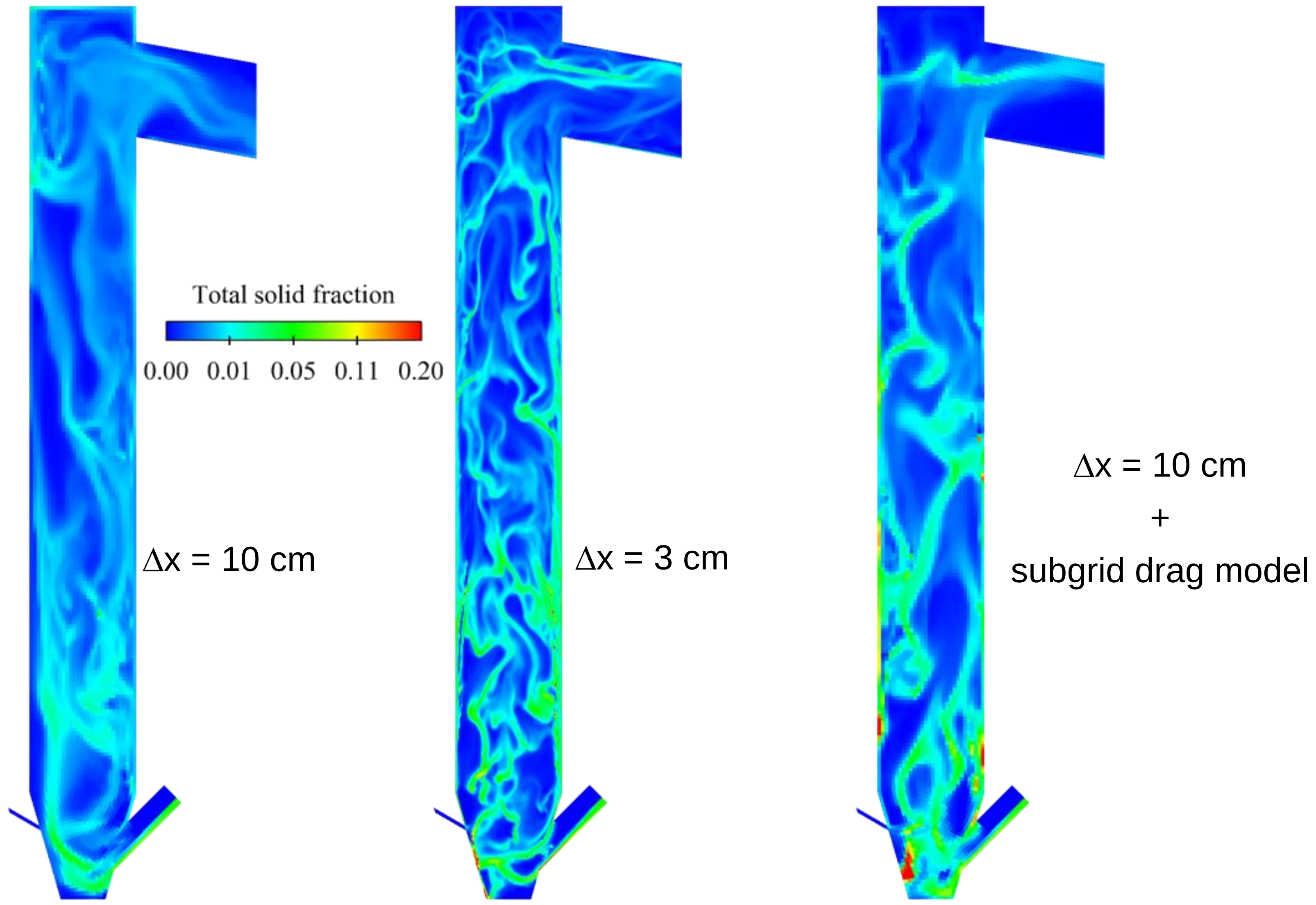
K_{ij}: Dynamic model (Germano et al., 1991)

$f(\Delta/\Delta_c)h(\overline{\alpha}_p)\widetilde{V}_{r,j}$: From *a priori* analysis

Prediction of solid mass flux

Influence of the mesh refinement:

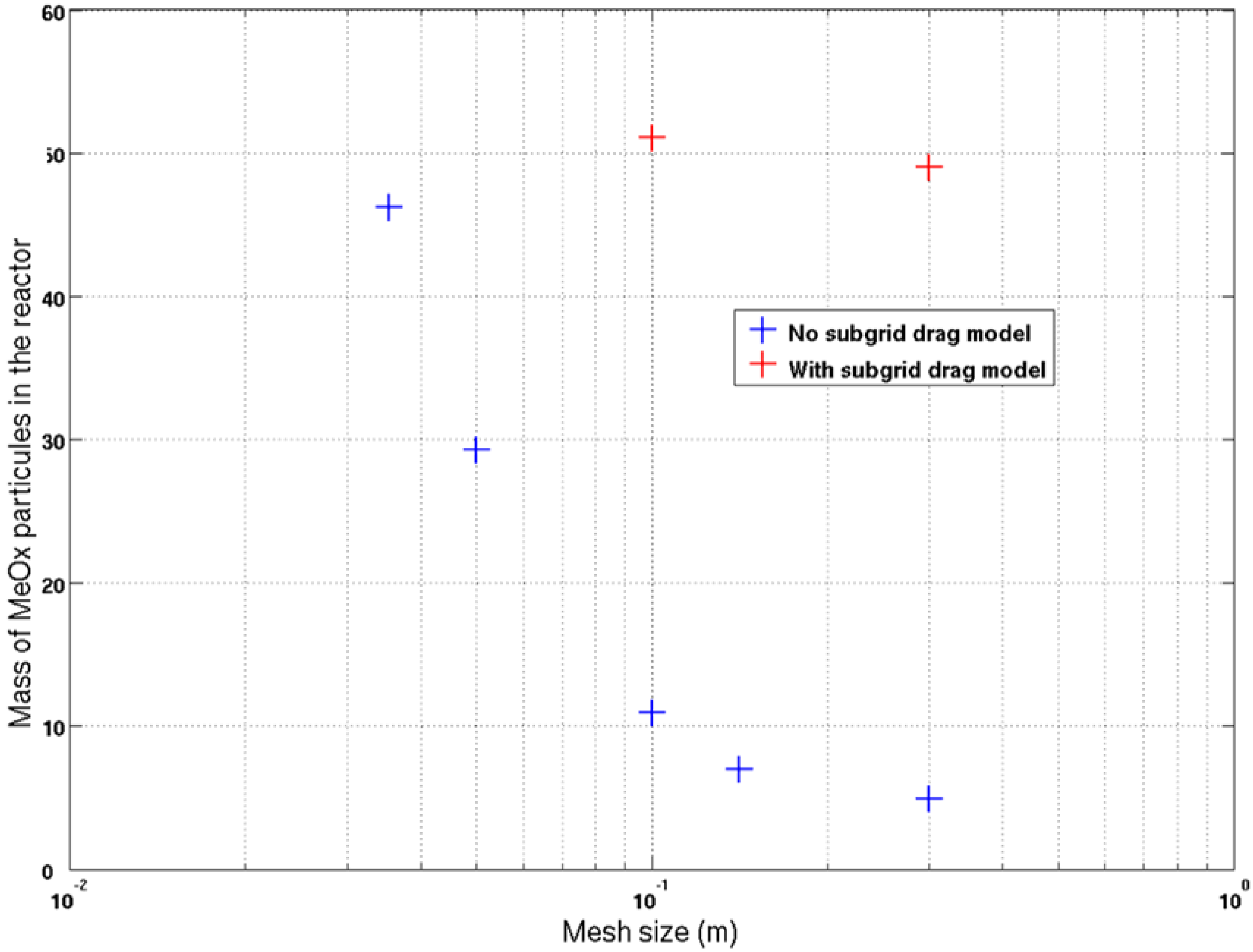
A posteriori test of the drag subgrid model on a circulating fluidized bed



Prediction of solid mass flux

Influence of the mesh refinement:

A posteriori test of the drag subgrid model on a circulating fluidized bed



Prediction of solid mass flux

	Total inventory	**Outlet solid mass flux**
EXP 1	260 kg	5.1 kg/s
EXP 2	188 kg	6.2 kg/s
EXP 3	174 kg	5.5 kg/s
EXP 4	151 kg	6.0 kg/s
NEPTUNE_CFD	260 kg	6.5 kg/s

In the range studied, the inventory of solid has no major influence on the circulating mass flux.

The simulation slightly overestimates the circulating mass flux.

Influence of the amount of fines on the recirculation of coarse particles

Gas phase: $T = 50°C$, $P = 1\ bar$

$V_{fluidization} = 3.8 m/s$ $\rho_g = 1.09\ kg/m^3$ $\mu_g = 1.9810^{-5}\ Pa \cdot s$

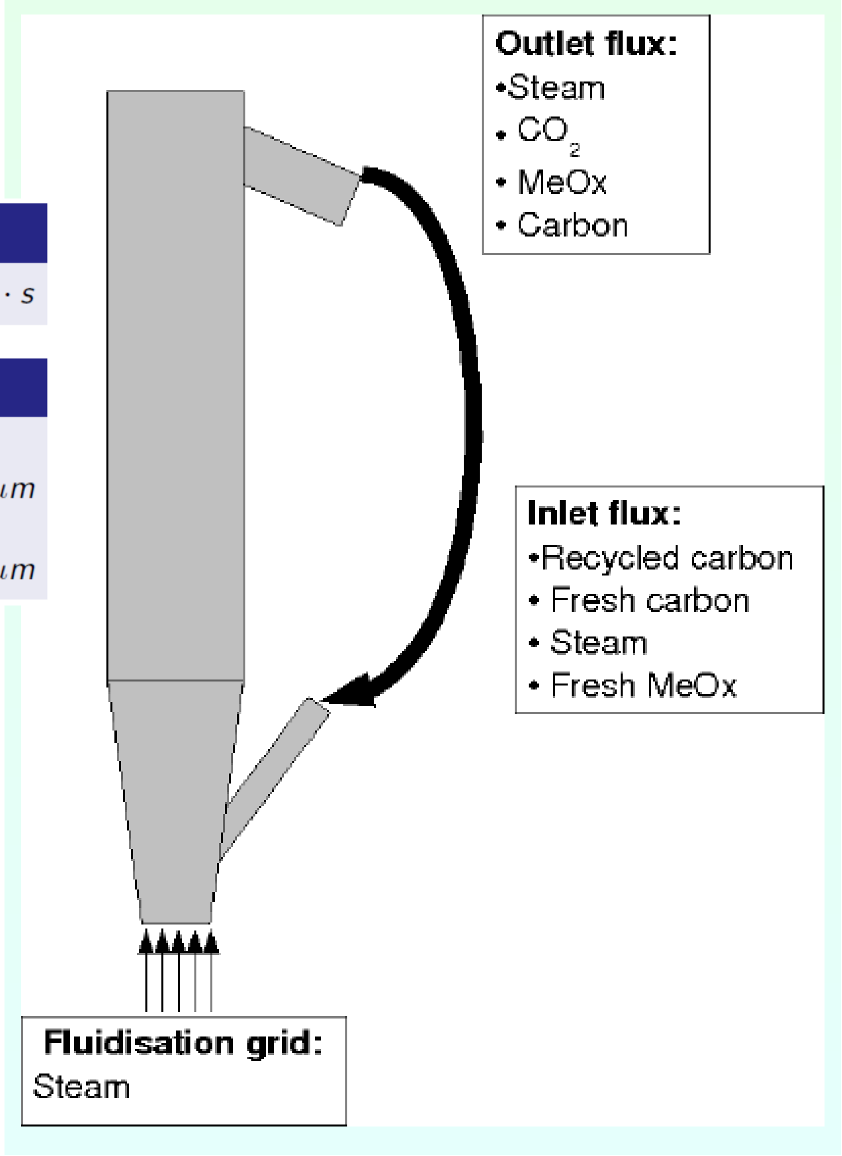
Solid phases

Ilmenite (for mono-solid and bi-solid experiments):

$V_t^{ilm} = 1.7 m/s$ $\rho_{ilm} = 4600\ kg/m^3$ $d_{50} = 160\ \mu m$

Alumina (for bi-solid experiments):

$V_t^{alu} = 0.14 m/s$ $\rho_{alu} = 1500\ kg/m^3$ $d_{50} = 60\ \mu m$



Influence of the amount of fines on the recirculation of coarse particles

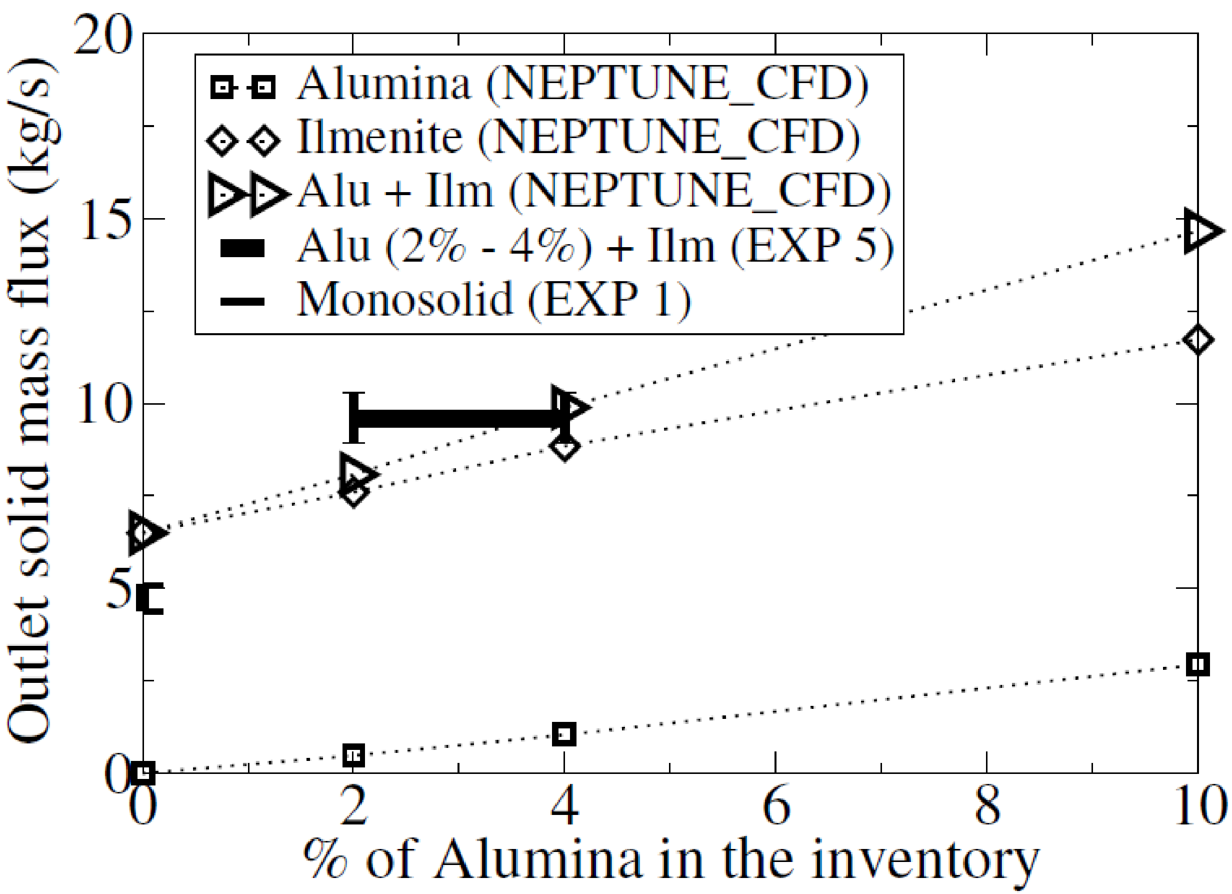
	Total inventory	% of Alumina	Ilmenite mass flux	Alumina mass flux	**Total mass flux**
EXPE 5	259 kg	2%-4%	-	-	9.6 kg/s
EXPE 6	193 kg	$\simeq$ 5%	-	-	9.5 kg/s
Alu 2%	259 kg	2%	7.6 kg/s	0.48 kg/s	8.1 kg/s
Alu 4%	264.5 kg	4%	8.85 kg/s	1.04 kg/s	9.9 kg/s
Alu 10%	259 kg	10%	11.73 kg/s	2.94 kg/s	14.7 kg/s

Polydisperse Eulerian approach prediction is satisfactory

The simulations showed that multiplying the ratio of alumina by:

- a factor 2 increases the circulation of ilmenite of 16%
- a factor 5 increases the circulation of ilmenite of 54%

Influence of the amount of fines on the recirculation of coarse particles

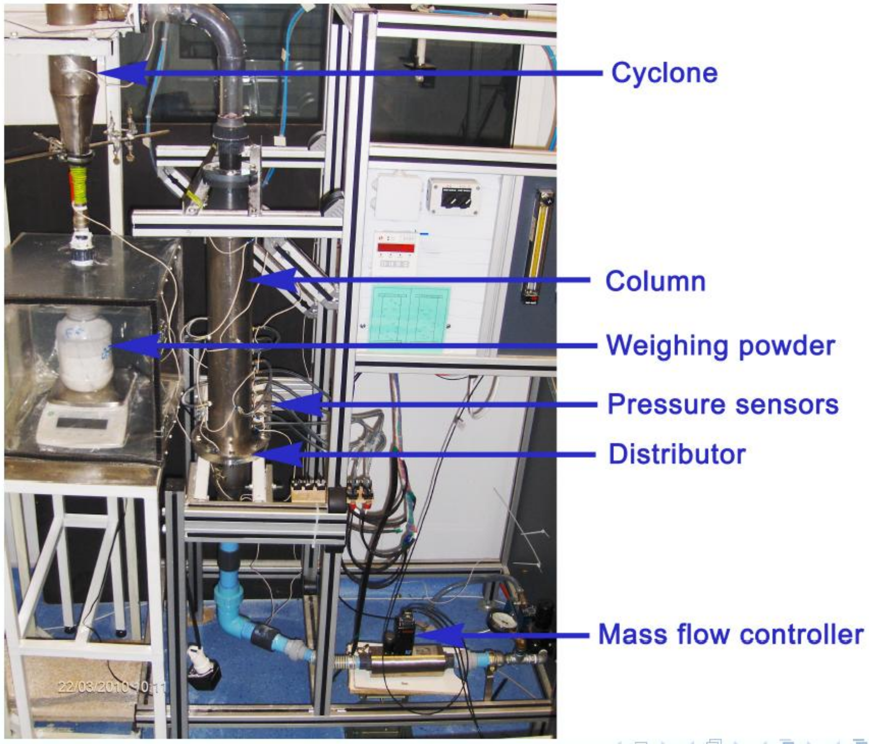


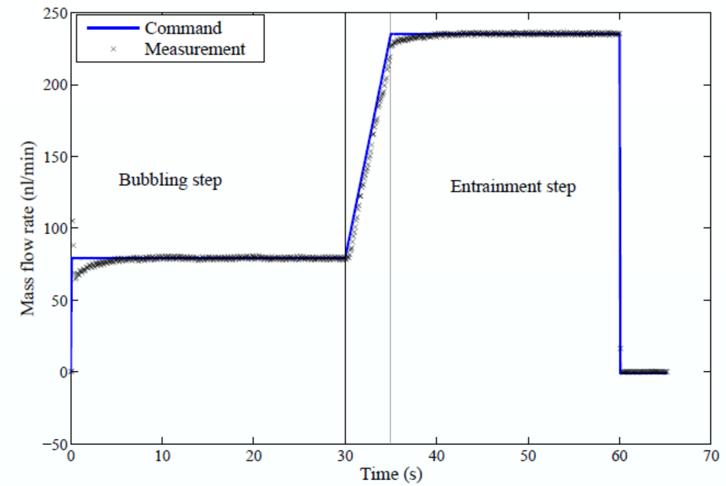
Ilmenite and alumina mass fluxes are increasing linearly with increasing the ratio of alumina

Influence of the amount of fines on the recirculation of coarse particles

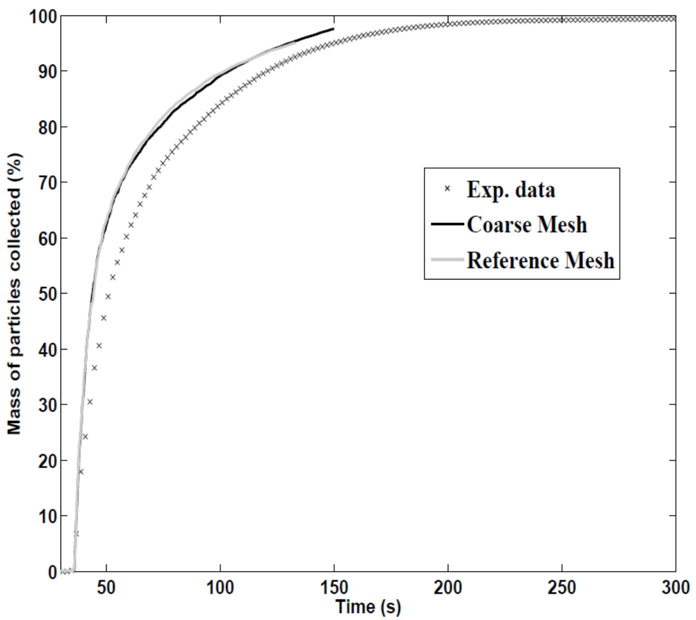
Model experiment:

Ansart et al., CFB-10, 2011





Particle properties	**Fine**	**Coarse**
Density $(\mathrm{kg/m^3})$	2470	2470
Geldart Group	A/B	B
Mean diameter d_{50} $(\mu\mathrm{m})$	84	213
Span$=\frac{d_{90}-d_{10}}{d_{50}}$	0.38	0.414
v_t $(\mathrm{m \cdot s^{-1}})$	0.41	1.51



Influence of the local gas production

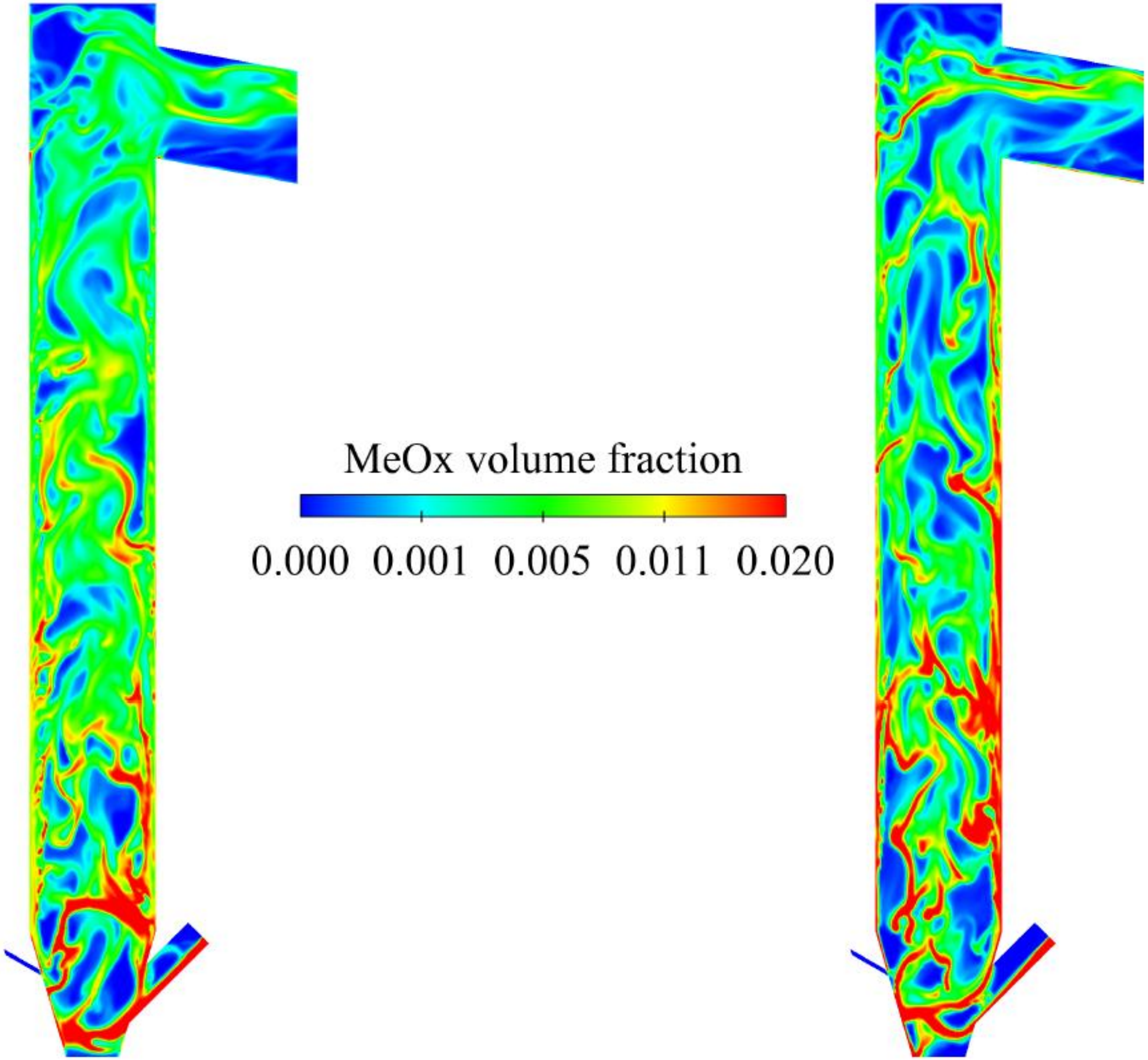
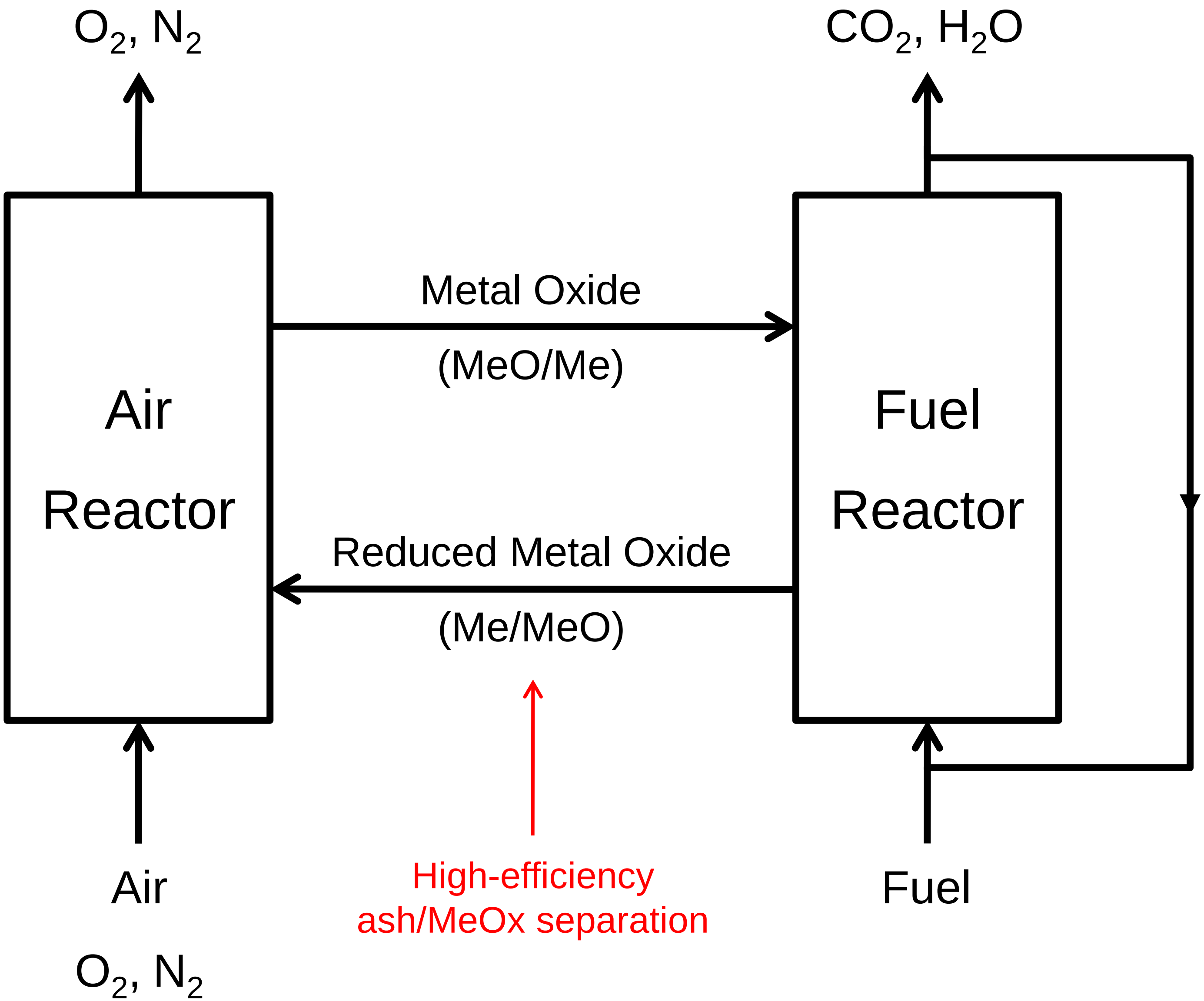


Figure 14: MeOx volume fraction in the reactor with reaction (left side) and without reaction (right side)

Ash/MeOx separation



Conclusions

→Hydrodynamic of fuel reactor:

- Numerical simulation A-type particles fluidization (in polydisperse case ?)
- Influence of the amount of fines on the recirculation of coarse particles
- Prediction of the local gas prediction (effect on the hydrodynamic)

→Separation : Ash/MeOx separation

→Numerical simulation of medium-scale pilot (1MW), delay in experiments conducted at Darmstadt University

→Coupled system: Numerical simulation of coupled air and fuel reactors