Driving Innovation ♦ Delivering Results

# Development of a novel CFD model for large-scale REE extraction process

**Project**: Rare Earth Elements Extraction from Coal By-Products

**Period of performance:** (Oct 2015 – Present)


**2017 Rare Earth Elements Portfolio Review**
**03/22/2017**


**Liqiang Lu (NETL/ORISE)**
**Sofiane Benyahia (NETL)**

# Project Objective and Goals

- **Develop computational tools to help design/optimize/scale-up processes of REE leaching with Ammonium sulfate from coal byproducts (clay) :**
  - Implement and validate an REE extraction reaction mechanism in a computational fluid dynamic (CFD) code.
  - Develop a novel and fast CFD model based on discrete particle method to simulate large-scale REE extraction reactors.
  - Validate the above CFD model for hydrodynamics, heat/mass transfer, and REE reaction kinetics with available experimental data from open literature.
  - Application of the above model to 3D counter-current continuous fluidized bed leaching reactor to optimize REE concentration in product.
  - Study the effects of scaling the leaching reactor from small to medium to large-scale fluidized beds.

# Outline

- Major CFD development: Coarse graining and hard-sphere models
- Model validation for accuracy and speed of execution
  - Particle packing for hard-sphere model
  - Bubbling fluidized bed case
  - Particle transport in the riser section of circulating fluidized bed
  - Heat transfer in a fluidized bed
- Chemical reaction model implementation and validation for REE leaching process
  - Model development
  - Leaching in a bubbling fluidized bed
  - Leaching in a bed of particles mixed with an impeller
- 3D counter-current leaching application: scale-up from a lab-scale to an industrial-scale reactor

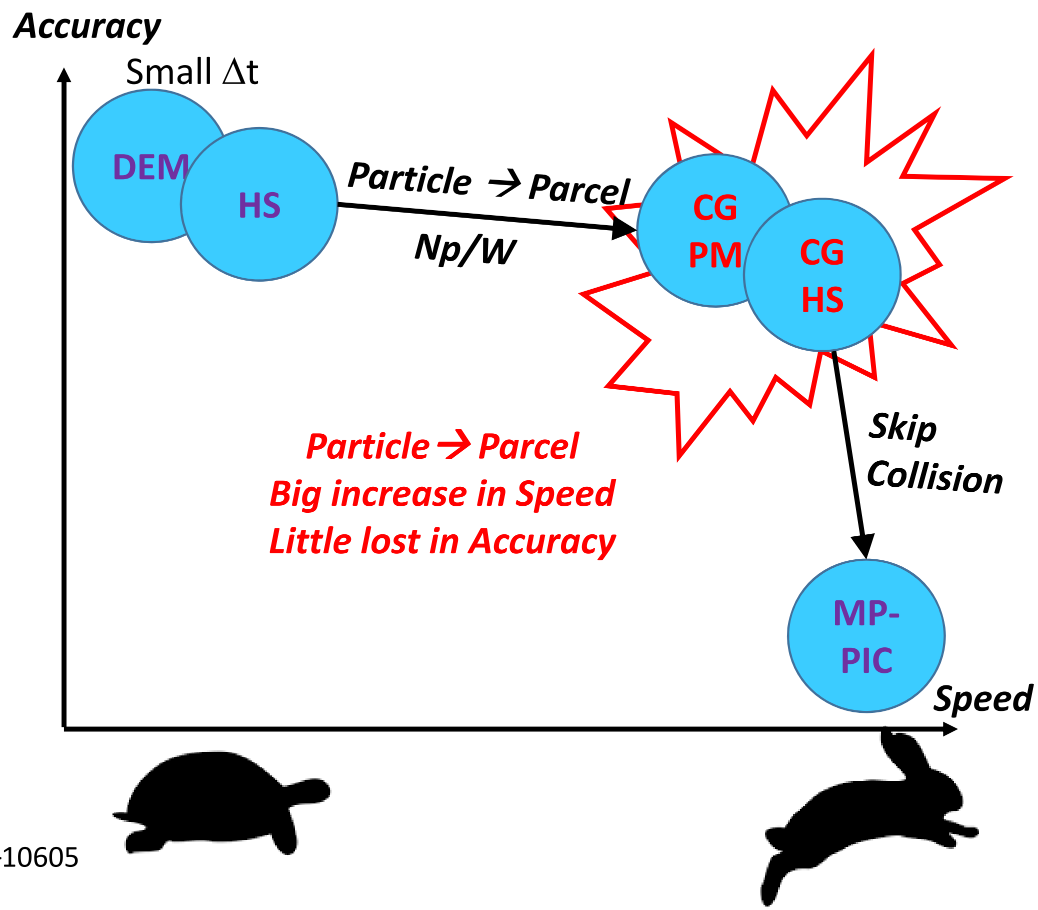
Core Technology Development

Technology Validation with Available Experimental Data

Technology Application to REE Process Optimization & Scale-up

**_Euler = CFD on Euler fixed Grids_**
- ***Strategy to solve incompressible NS equations (SIMPLE, PISO)***
- ***Linear Equation Solver***

**_Lagrange = Newton's Law of motion_**
- ***particle(DEM)/parcel (PIC)***
- ***Particle-particle forces***




**_CG is accurate because of our previous work:_**
- Benyahia, Galvin: Ind. Eng. Chem. Res. 2010, 49, 10588–10605
- Lu, Xu, Ge, et al. Chem. Eng. Sci. 2016, 155, 314-337.

# CFD code development: Coarse graining for Hydrodynamics and Chemical Reactions

Fluid phase equations

| | |
|---|---|
| Fluid phase continuity: | $\frac{\partial(\varepsilon_f \rho_f)}{\partial t} + (\nabla \cdot \varepsilon_f \rho_f \mathbf{u}_f) = \sum_{n=1}^{N_f} R_{fn}$ |
| Fluid phase momentum: | $\frac{\partial(\rho_f \varepsilon_f \mathbf{v}_f)}{\partial t} + \nabla \cdot (\rho_f \varepsilon_f \mathbf{v}_f \mathbf{v}_f) = -\varepsilon_f \nabla P_f + \nabla \cdot (\boldsymbol{\tau}_f) - \mathbf{I} + \varepsilon_f \rho_f \mathbf{g}$ |
| Interphase momentum transport: | $\mathbf{I}^k = \frac{1}{v_k} \sum_{i=1}^{N_p} \frac{\beta^i \frac{1}{6}\pi d_p^3}{1-\varepsilon_f} W_p (\mathbf{v}_f(\mathbf{x}^i) - \mathbf{v}_p^i) K(\mathbf{x}^i, \mathbf{x}_k)$ |
| Species transport: | $\frac{\partial(\varepsilon_f \rho_f X_i)}{\partial t} + \nabla \cdot (\varepsilon_f \rho_f \mathbf{u}_f X_i) = \nabla \cdot (D_i \nabla X_i) + S_{Xi}$ |
| Constitute equations: | $R_{fn} = -\frac{1}{v_k} \sum_{i=1}^{N_k} W_i R_{fn}^i K(\mathbf{x}^i, \mathbf{x}^k) \quad Re_p = \frac{\varepsilon_f \rho_f \lvert \mathbf{v}_f - \mathbf{v}_p \rvert d_p}{\mu_f}$ |

Solid phase equations

$$m_p \frac{d\mathbf{v}_p}{dt} = \mathbf{F}_b + \mathbf{F}_{gp} + \mathbf{F}_{drag} + \mathbf{F}_c$$

$$\mathbf{F}_b = m_p \mathbf{g} \qquad \mathbf{F}_{gp} = -\nabla P_g \frac{\pi}{6} d_p^3$$

$$\mathbf{F}_{drag} = \frac{\beta}{1-\varepsilon_f}(\mathbf{u}_f - \mathbf{v}_p)\frac{\pi}{6} d_p^3$$

$$\mathbf{F}_c = \sum_{j=1, j\neq i}^{N} (\mathbf{F}_{ij}^n + \mathbf{F}_{ij}^t) / W$$

$$\mathbf{T} = \sum_{j=1, j\neq i}^{N} (L\mathbf{n} \times \mathbf{F}_{ij}^t) / W^{5/3}$$

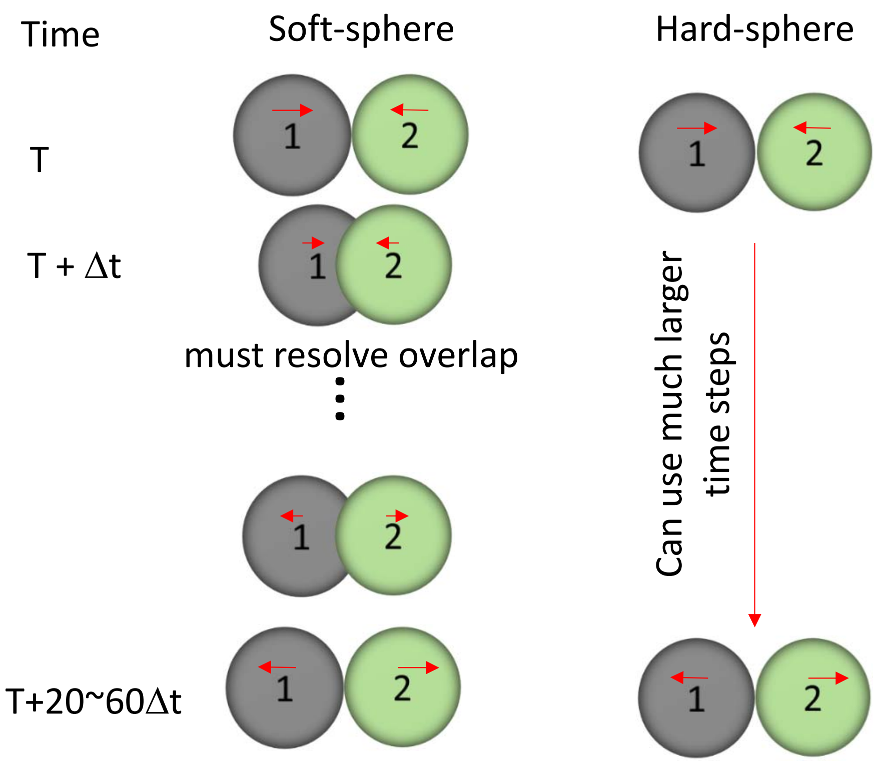
The only parameter in this model is: statistic weight *W*

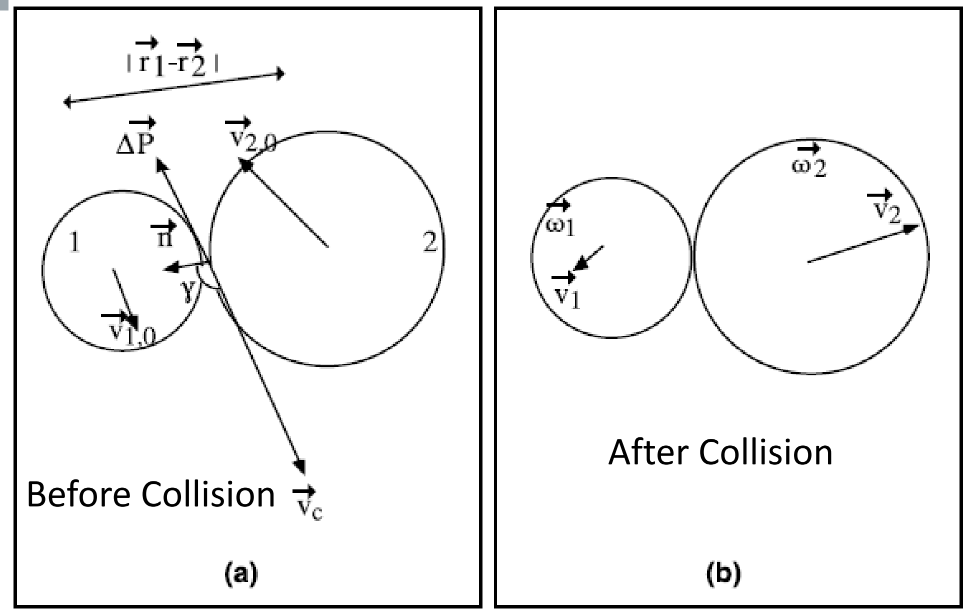
*Accuracy* — *W* — *Speed*

*DEM 1 100 200 …… ?*

- *Compromise between accuracy and speed*
- *Different system has different maximum W*
- *With more accurate interpolation method, Larger W can be used*

***Introducing a Novel Hard-Sphere Contact Model:***
***As Accurate as Soft-Sphere Model But much Faster!***

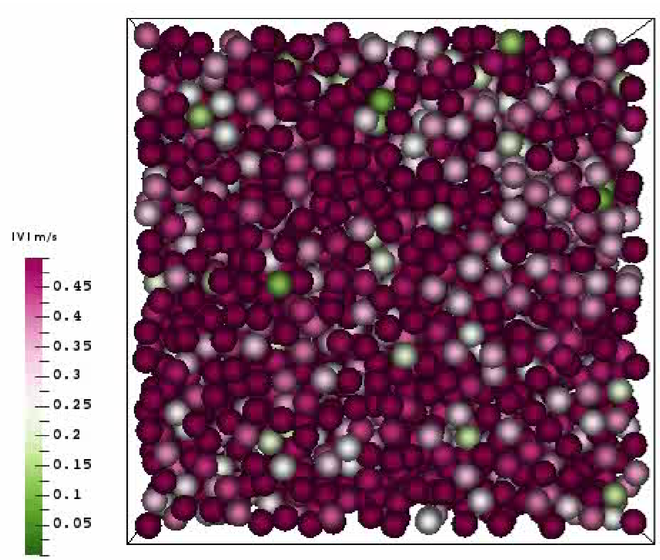


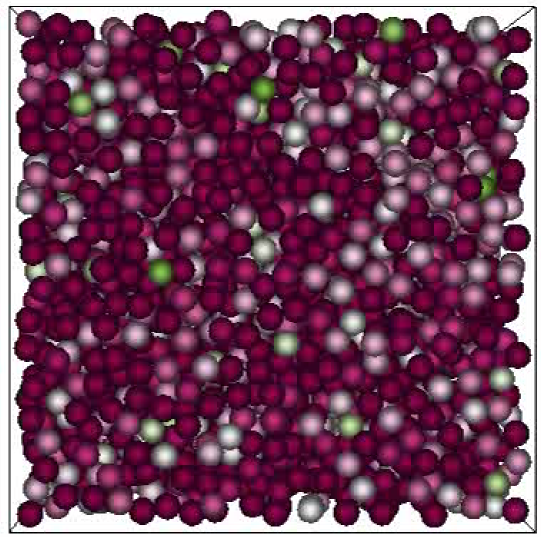

$$\mathbf{J}_t = -\max\left(\frac{2}{7} m_e (1+e_t)\left|\mathbf{v}_t\right|, \mu m_e (1+e_n)\left|\mathbf{v}_n\right|\right) \frac{\mathbf{v}_t}{\left|\mathbf{v}_t\right|}$$

$$\mathbf{v}_i = \mathbf{v}_i - \frac{\Delta \mathbf{P}}{m_i} \qquad \mathbf{v}_j = \mathbf{v}_j + \frac{\Delta \mathbf{P}}{m_j}$$

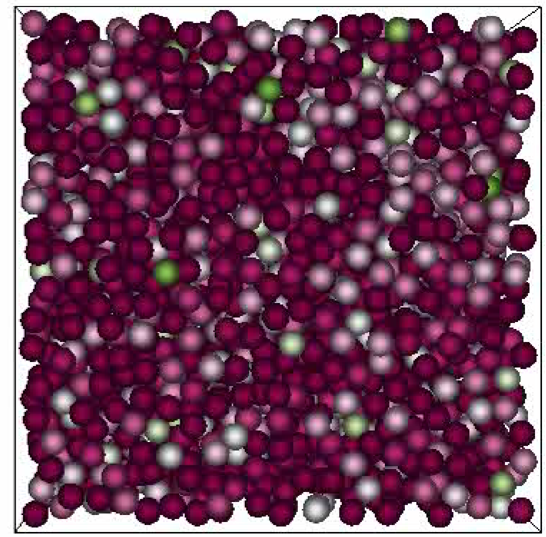
$$\boldsymbol{\omega}_i = \boldsymbol{\omega}_i - \frac{r_i \mathbf{n} \times \mathbf{J}_t}{I_i} \qquad \boldsymbol{\omega}_j = \boldsymbol{\omega}_j - \frac{r_j \mathbf{n} \times \mathbf{J}_t}{I_j}$$




Soft-sphere



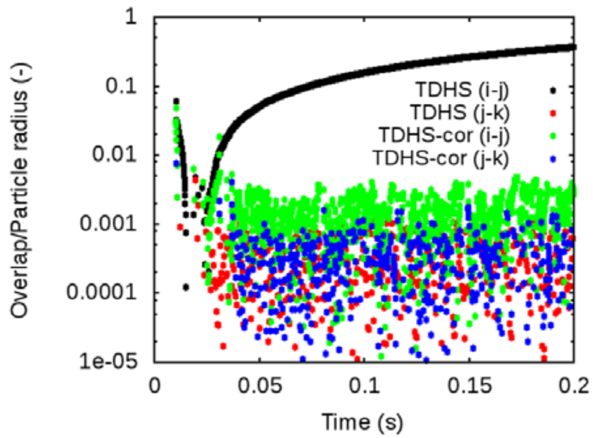
Hard-sphere-orginal

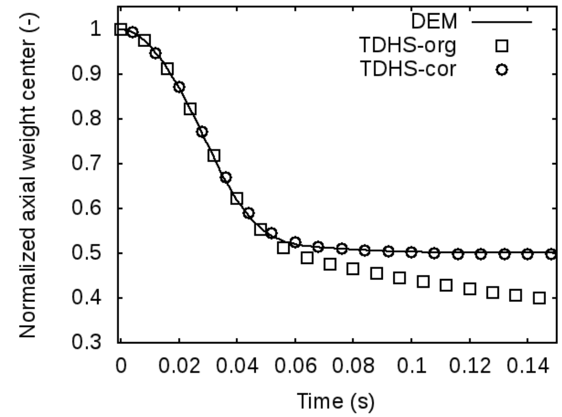


Hard-sphere-corrected

inelastic collapse of a granular assembly under gravity.

```
loop1: iter=1, corSteps and !noMoreCollide
    noMoreCollide = .true.
    forall1 particle i
        forall2 particle i's neighbor j
            if(iter==1)    goal(i,j) = 0
            if(r_ij v_n>0 && |r_ij|<r_i+r_j)
                If(iter==1)    goal(i,j) = -v_n
                Δv = -k_overlap δ_n + k_velocity(goal(i,j) - |v_n|)
                v_i= v_i − nΔv
                v_j= v_j + nΔv
                noMoreCollide = .false.
            endif
        endforall2
    endforall1
endloop1
```

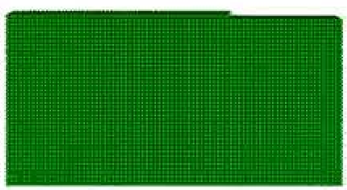


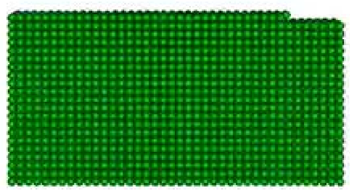

This correction to original hard-sphere model is novel and will allow simulation of fluidized beds with increased simulation speed.

***Combining coarse-graining and hard-sphere methods: Gaining at least 3-4 orders of magnitude speed-up!***

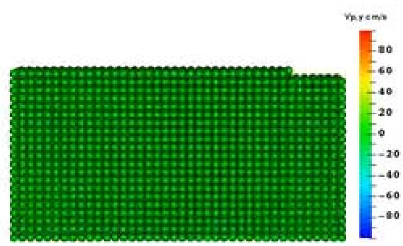
# *Coarse grained hard sphere : validation through a small BFB*



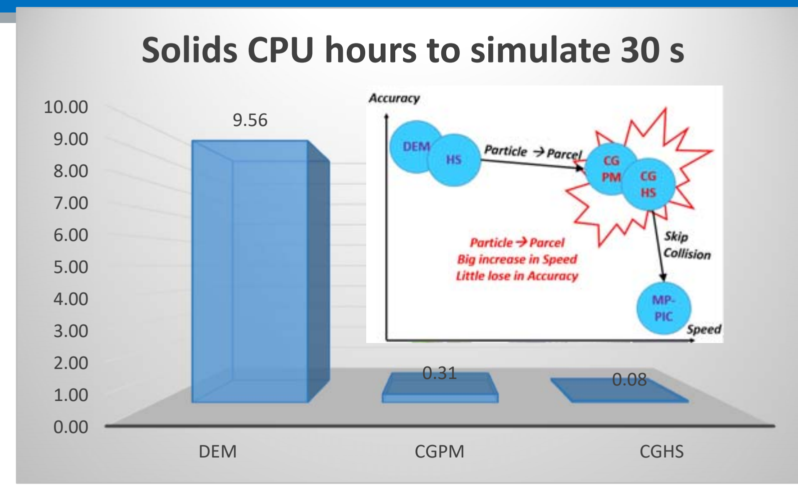
DEM
$\Delta t = 1.79 \times 10^{-5}$ s

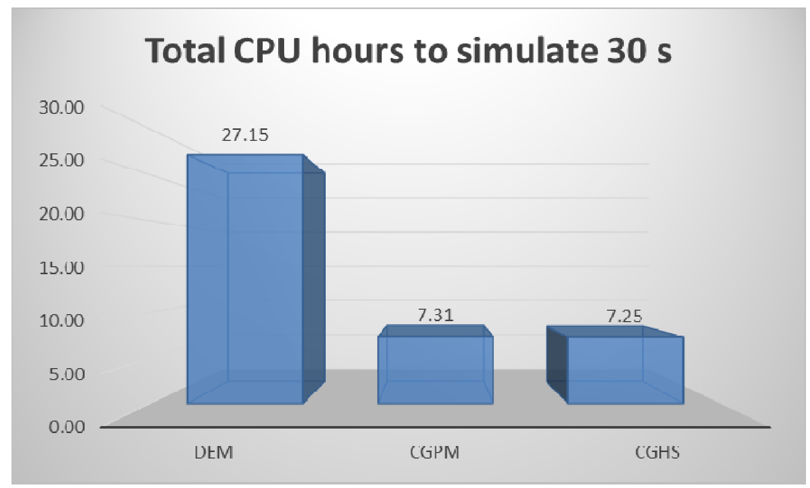


CGPM-W8
$\Delta t = 5.05 \times 10^{-5}$ s

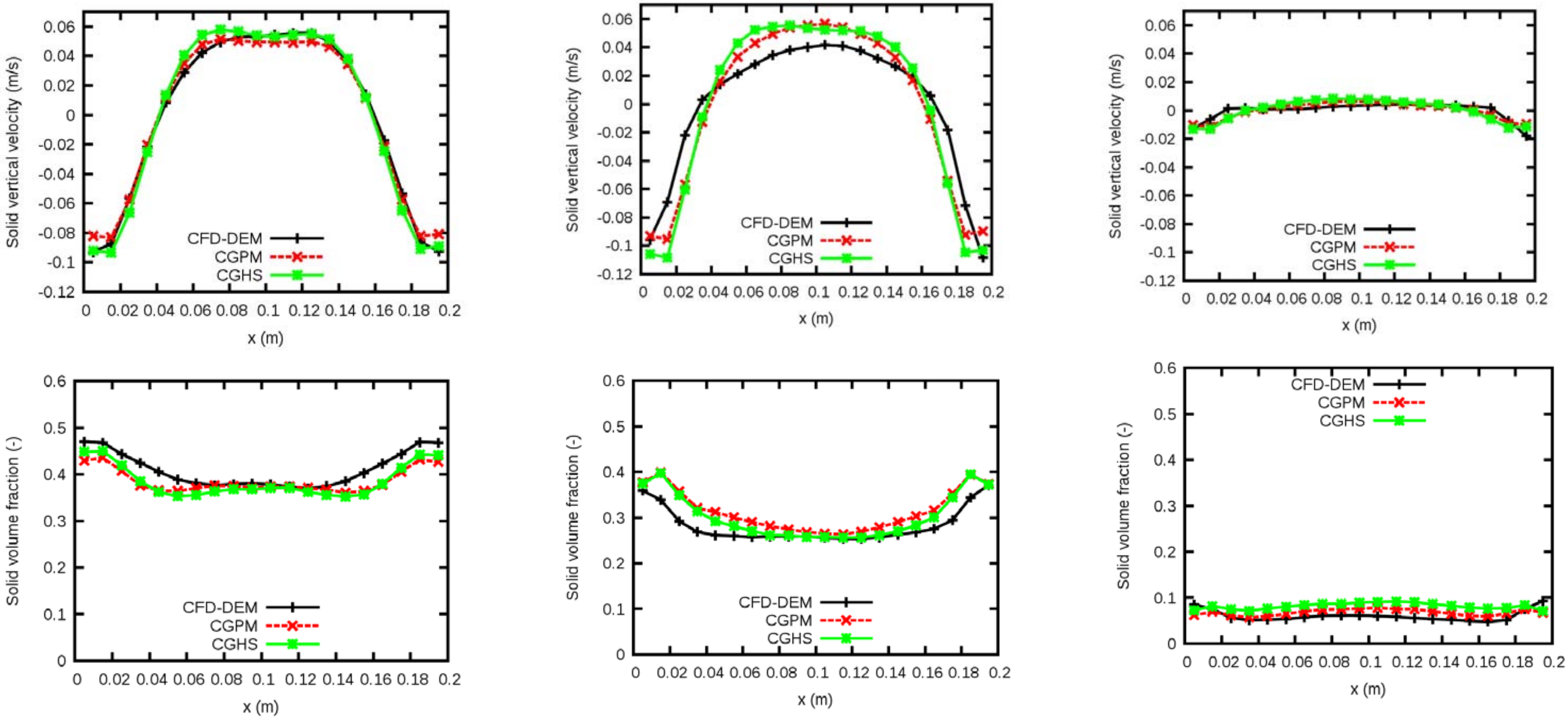


CGHS-W8
$\Delta t = 1 \times 10^{-3}$ s

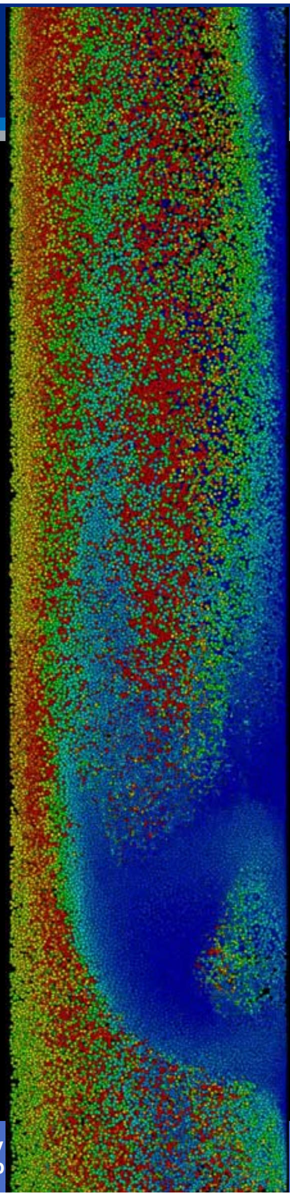


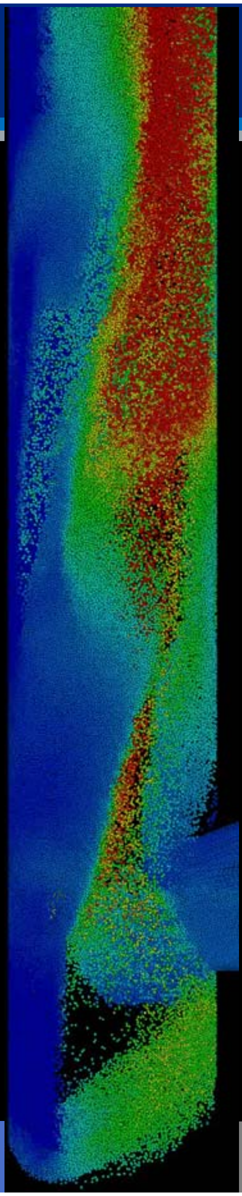



# *Coarse grained hard sphere : validation through a small BFB*



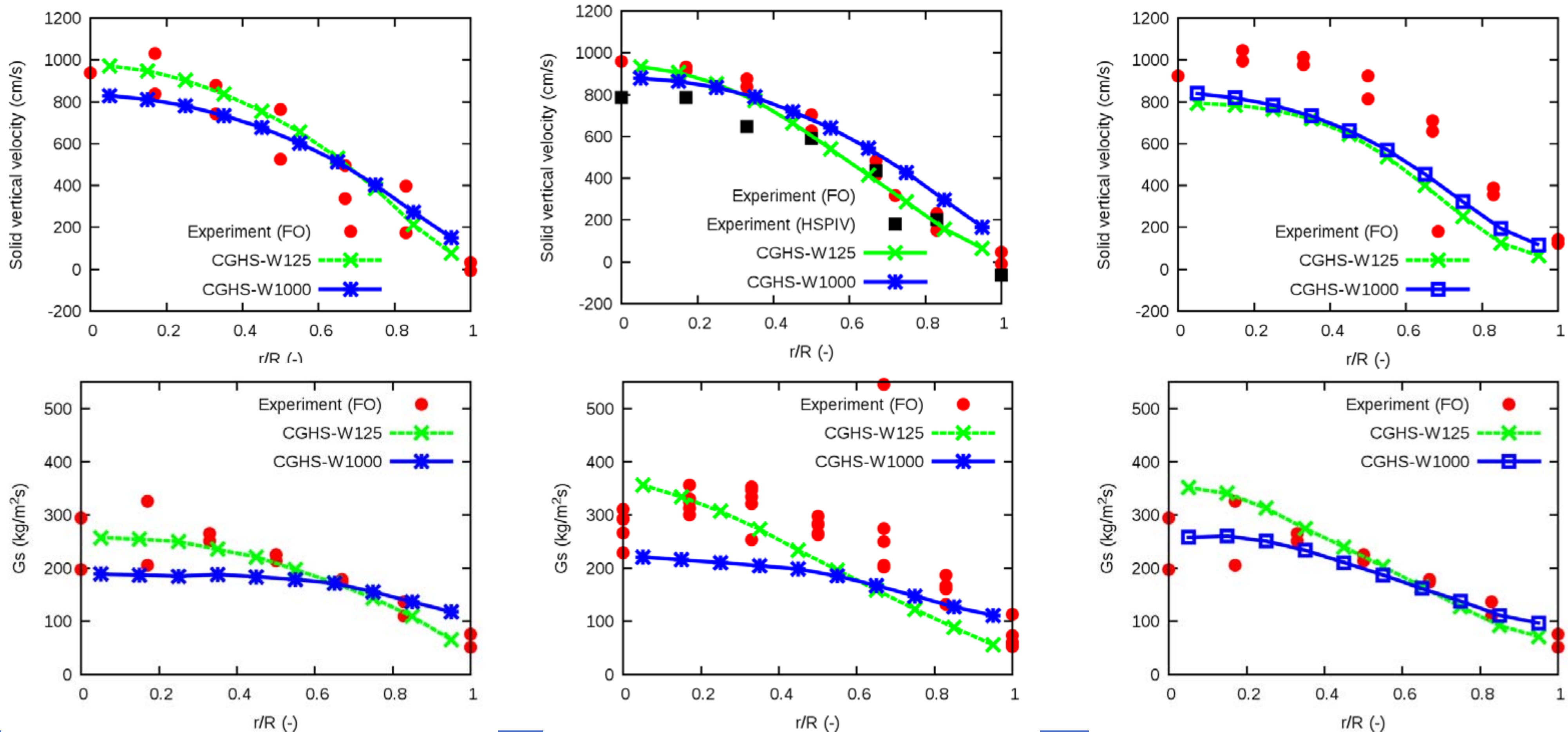
# CGHS: B22 riser



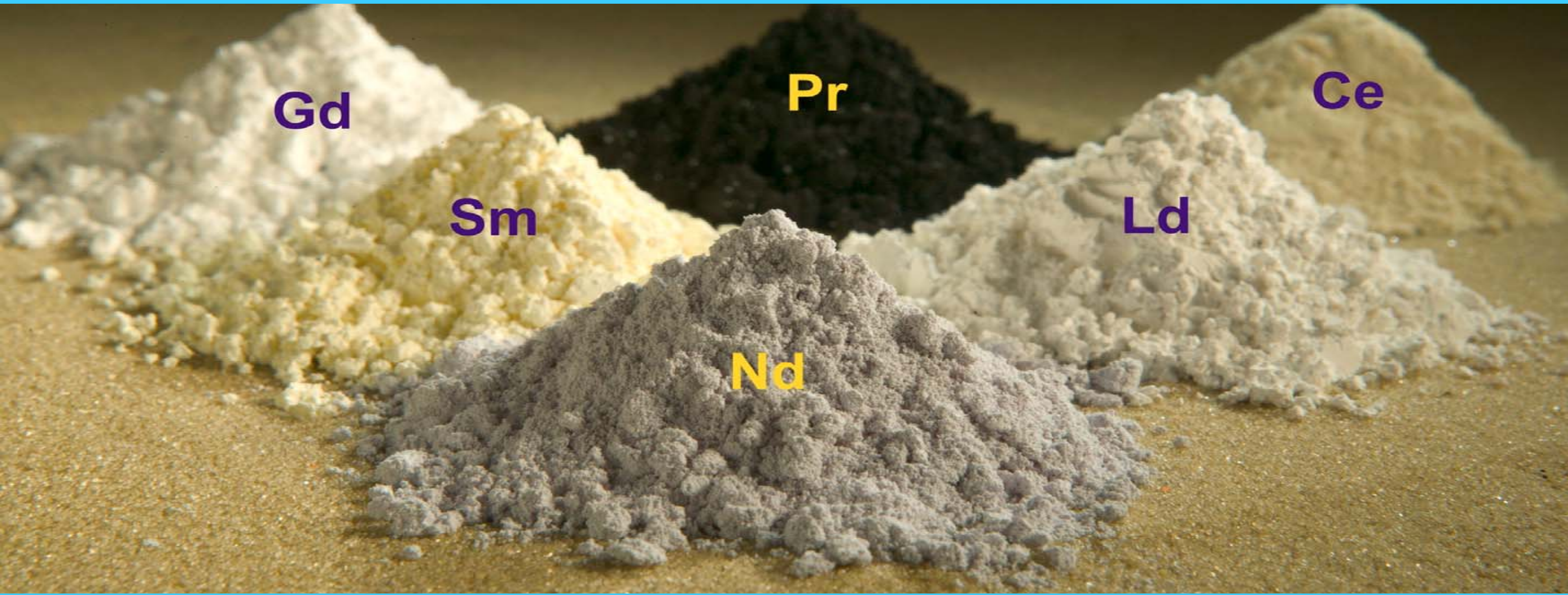


# CGHS: B22 riser

H = 6.23 m    H = 8.88 m    H = 13.33 m

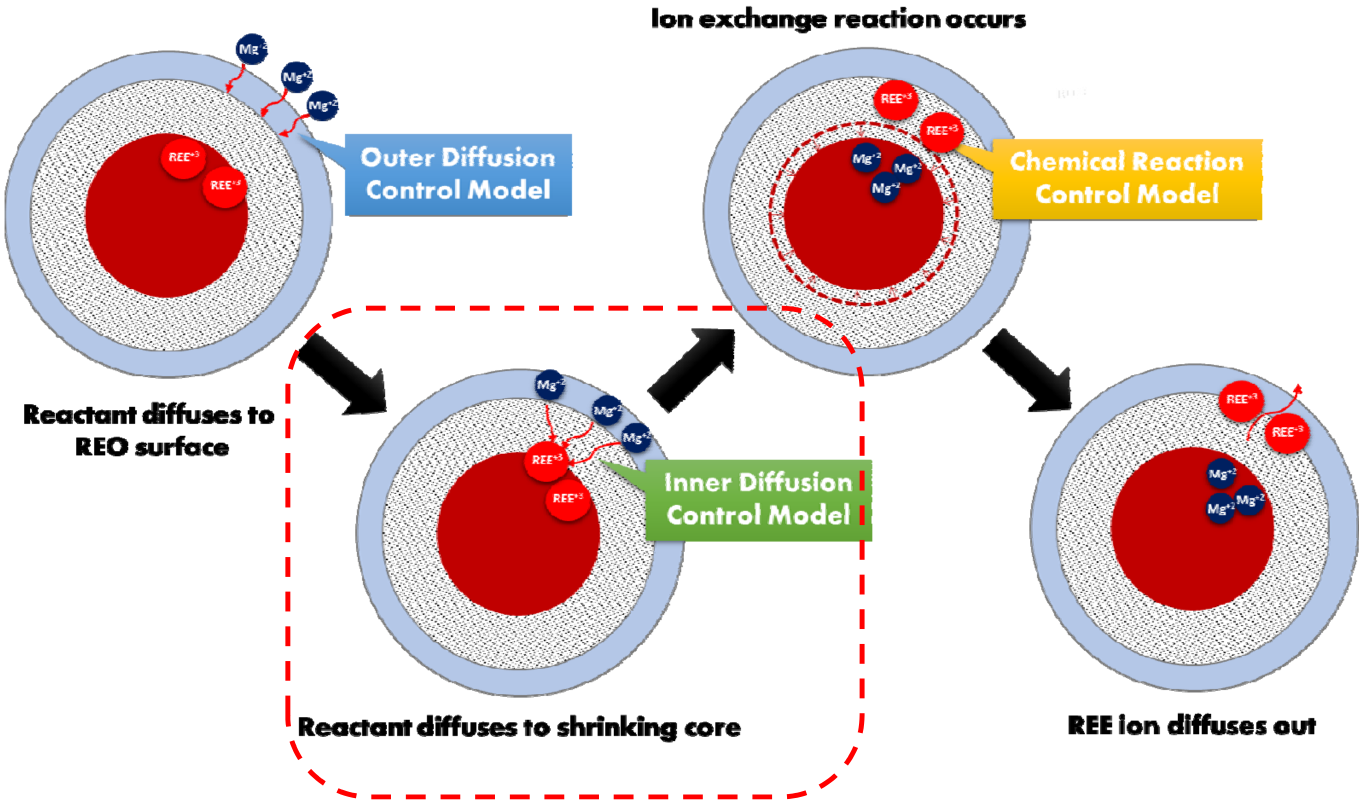


# MFIX-CGPM:
## *Hydrodynamics and Chemical Reactions*




## Rare Earth Element Leaching

$$\left\{Al_4\left[Si_4O_{10}\right](OH)_8\right\}_m^- \cdot 2nRE_{(s)}^{3+} + 3nMg_{(f)}^{2+} \rightarrow \left\{Al_4\left[Si_4O_{10}\right](OH)_8\right\}_m^- \cdot 3nMg_{(s)}^{2+} + 2nRE_{(f)}^{3+}$$



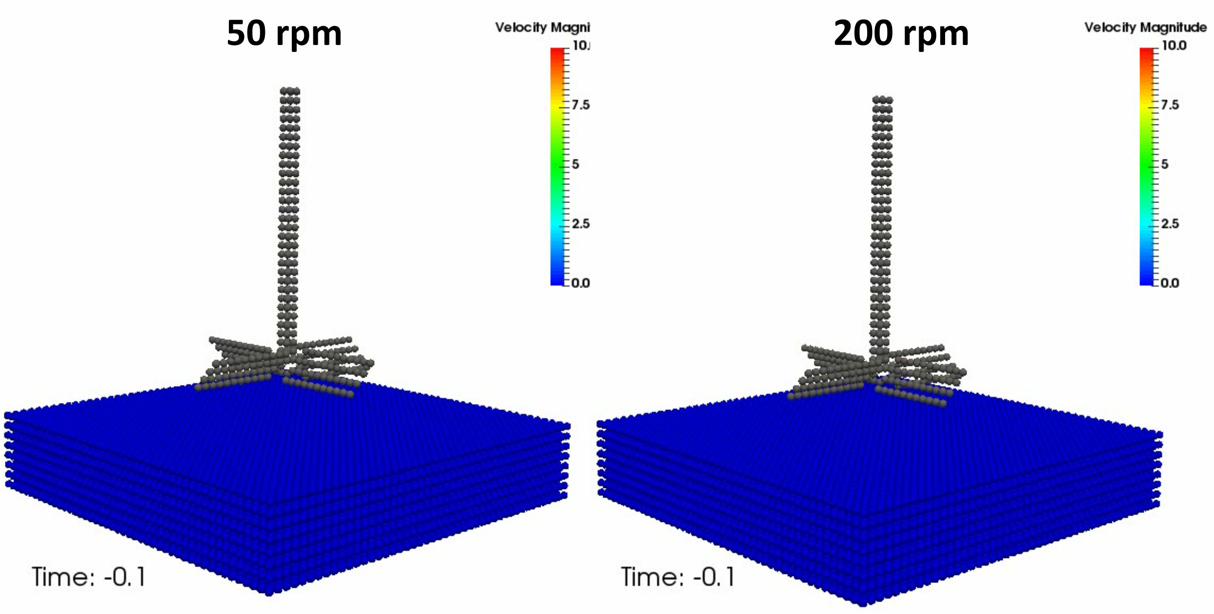

$$1-\frac{2}{3}\alpha-(1-\alpha)^{2/3}=kt$$

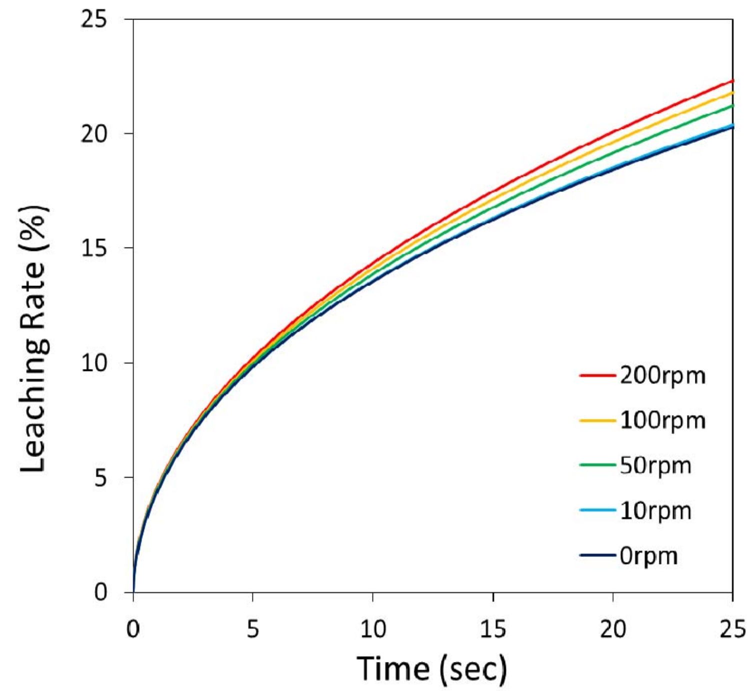
$$\alpha=1-\left(\frac{r_c}{R_p}\right)^3$$

$$k_{\exp}(1/\min)=\frac{0.011}{R_p(mm)^{1.217}}\exp\left(-\frac{9480}{RT}\right)$$

$$k(1/\min)=\frac{C_{Mg}}{C_{Mg,\exp}}k_{\exp}$$

Yoo, K.; Lu, L.; Benyahia, S., Modeling and simulation of rare earth element extraction by ion exchange reaction in batch reactor. *Internal report*.

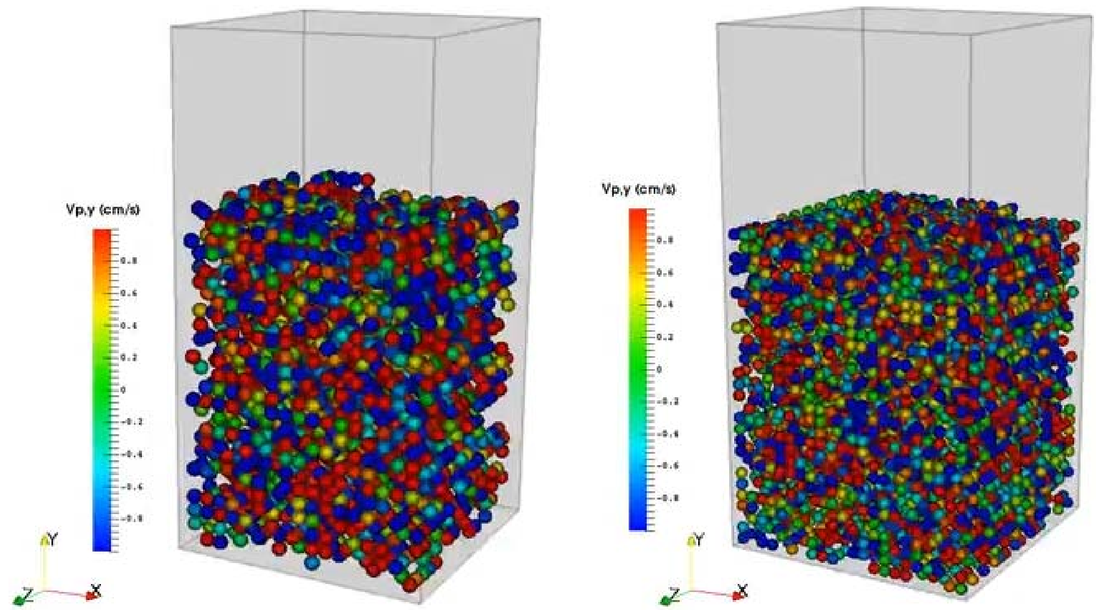


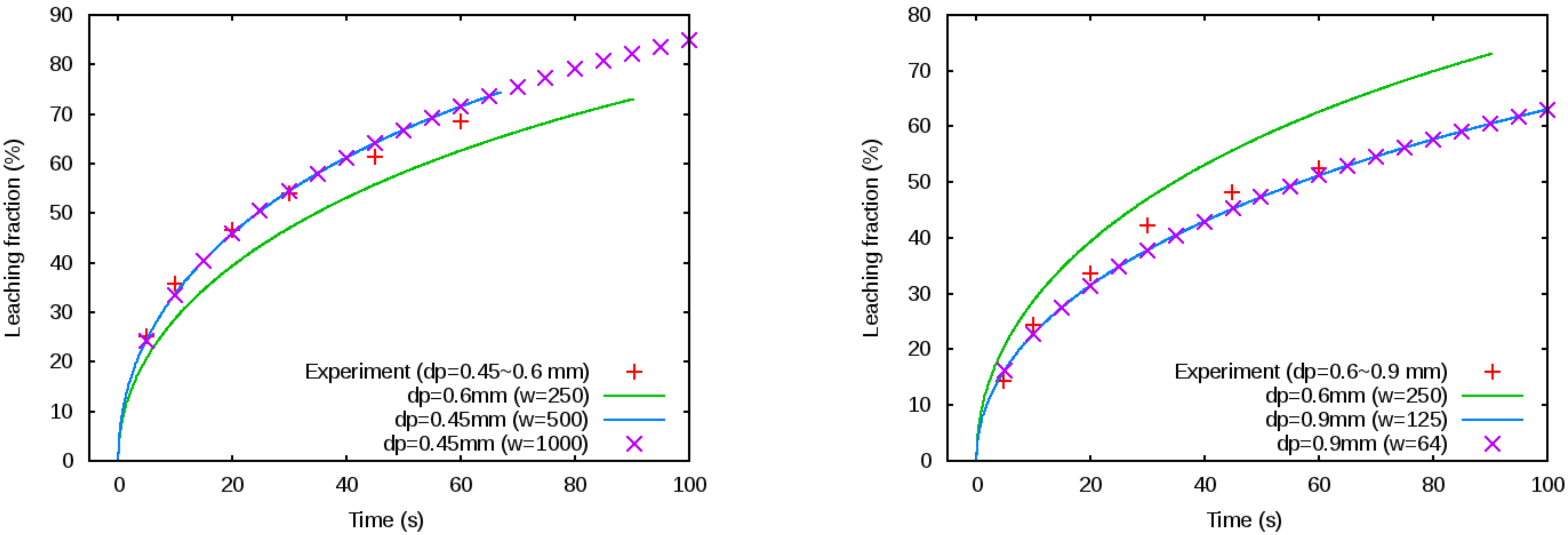

- For 50 rpm case, most REO particles pile up at the bottom of reactor and
- At higher rotating speed of 200 rpm, most REO particles are fluidized
- higher leaching rate is obtained at faster rotating speed.

| | Properties | value |
|---|---|---|
| **Particle** | Diameter, $d_p$ (cm) | 0.045, 0.06, 0.09 |
| | Density, $\rho_p$ (g/cm$^3$) | 1.34 |
| | Particle components mass fraction, REE/$NH_4$/Ash | 0.00102/0.0/0.99898 |
| | Spring constant, $k_n$ (dyne/cm) | 1.0x10$^5$ |
| | Restitution coefficient, $e$ | 0.7 |
| | Friction coefficient, $\mu$ | 0.1 |
| | Time step, $\Delta t$ (s) | Varied |
| Liquid | Density, $\rho_l$ (g/cm$^3$) | 1.0 |
| | Viscosity, $\mu_l$ (g/cm/s) | 0.01 |
| | Initial liquid components mass fraction, REE/$NH_4$/$SO_4$/$H_2O$ | 0.0/0.002730/0.007346/0.989924 |
| | Time step, $\Delta t$ (s) | Varied |
| | | |

*Experiment:*
$d_{p1}$ = 0.045~0.060 cm
$d_{p2}$ = 0.060~0.900 cm




$d_p$ = 0.045cm, with $W$=1000(left) and $W$=500(right)



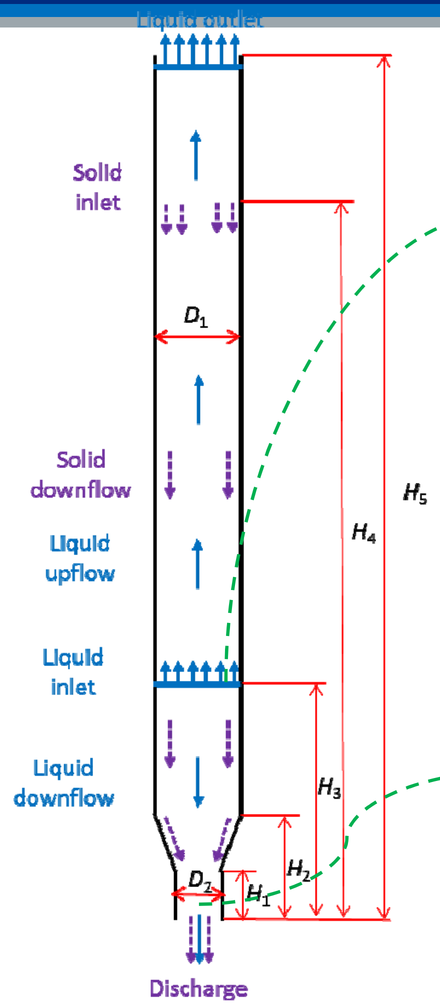

The simulation result compares well with experimental data

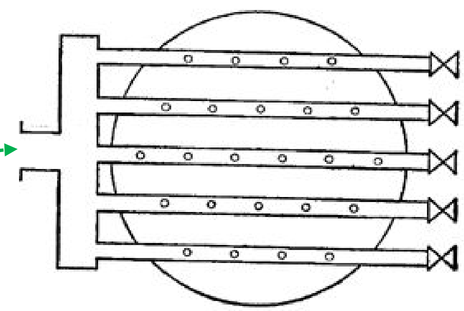
The result is insensitive to statistic weight

The system can be treated as monodisperse and corresponding diameter is 0.45 mm and 0.9 mm.

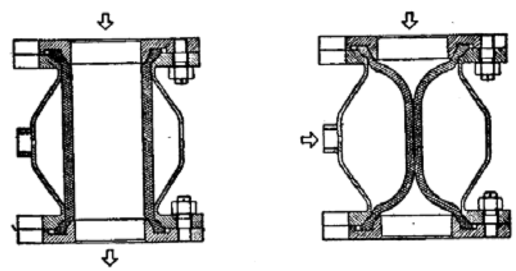
Xiao, Y.-f., Chen, Y.-y., Feng, Z.-y., Huang, X.-w., Huang, L., Long, Z.-q., Cui, D.-l., 2015. Leaching characteristics of ion-adsorption type rare earths ore with magnesium sulfate. Transactions of Nonferrous Metals Society of China 25, 3784-3790

# *Counter-current fluidized bed: geometry and operating conditions*



Simulated with point source



Valve open Valve closed

Simulated with pressure drop in porous medium

*Varied operating condition & reactor geometry*

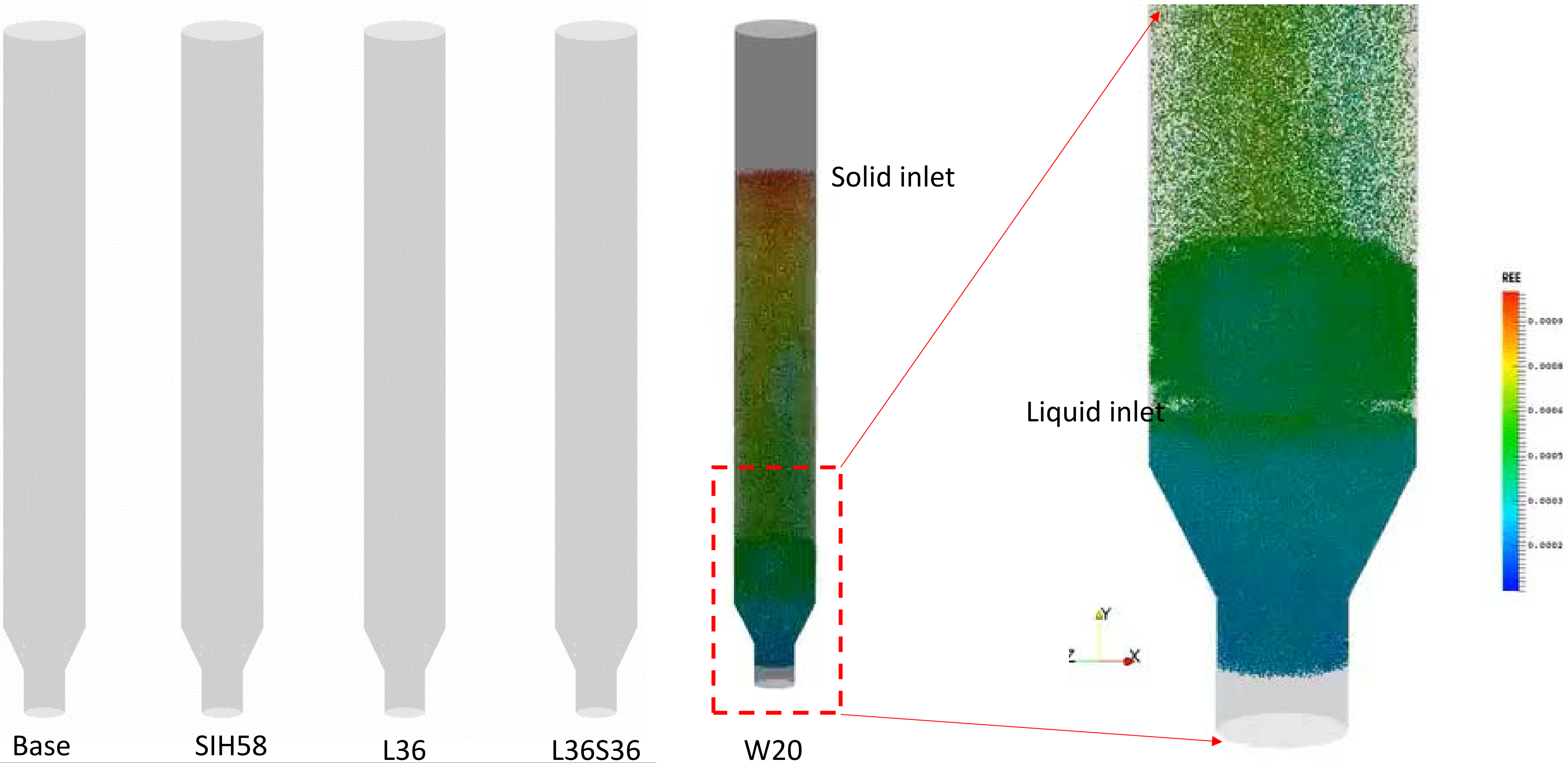
| Case | Base | Recycle50 | Recycle80 | RT100 | SIH58 | L36 | L36S3 |
|---|---|---|---|---|---|---|---|
| Liquid inlet flow rate, L (kg/h) | 72 | 72 | 72 | 72 | 72 | **36** | **36** |
| Solid inlet flow rate, S (kg/h) | 18.41 | 18.41 | 18.41 | 18.41 | 18.41 | 18.41 | **36.8** |
| Liquid recycle fraction, (%) | 20 | **50** | **80** | 20 | 20 | 20 | 20 |
| Residence time, (s) | 80 | 80 | 80 | **100** | **100** | 80 | 80 |
| Diameter, $D_1$ (cm) | 8 | 8 | 8 | 8 | 8 | 8 | 8 |
| Diameter, $D_2$ (cm) | 4 | 4 | 4 | 4 | 4 | 4 | 4 |
| Height, $H_1$ (cm) | 4 | 4 | 4 | 4 | 4 | 4 | 4 |
| Height, $H_2$ (cm) | 8 | 8 | 8 | 8 | 8 | 8 | 8 |
| Height, $H_3$ (cm) | 10 | 10 | 10 | 10 | 10 | 10 | 10 |
| Height, $H_4$ (cm) | 48 | 48 | 48 | 48 | **58** | 48 | 48 |
| Height, $H_5$ (cm) | 64 | 64 | 64 | 64 | 64 | 64 | 64 |

❖ Leachate recycling

❖ Residence time & inlet position

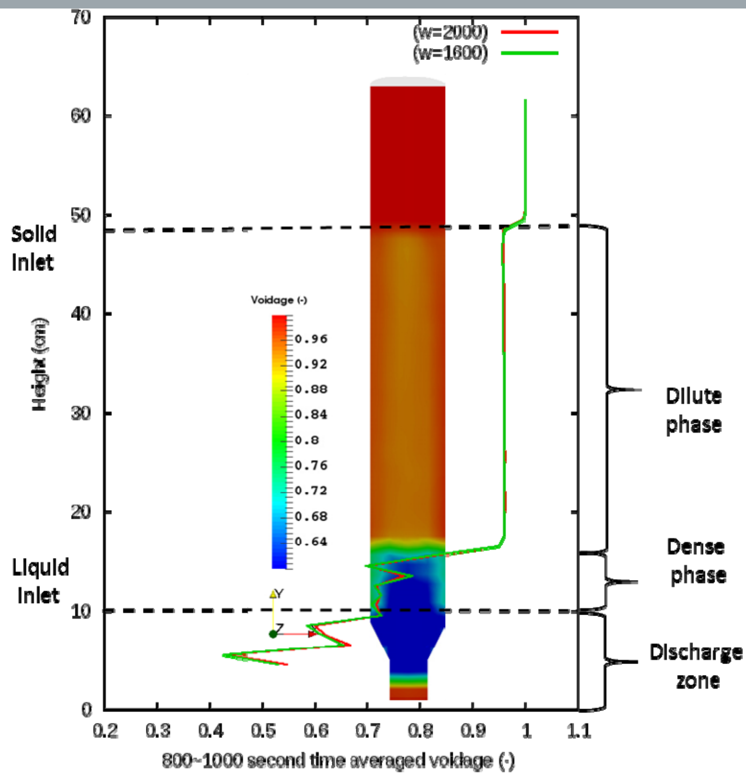
❖ Decreasing the L:S ratio

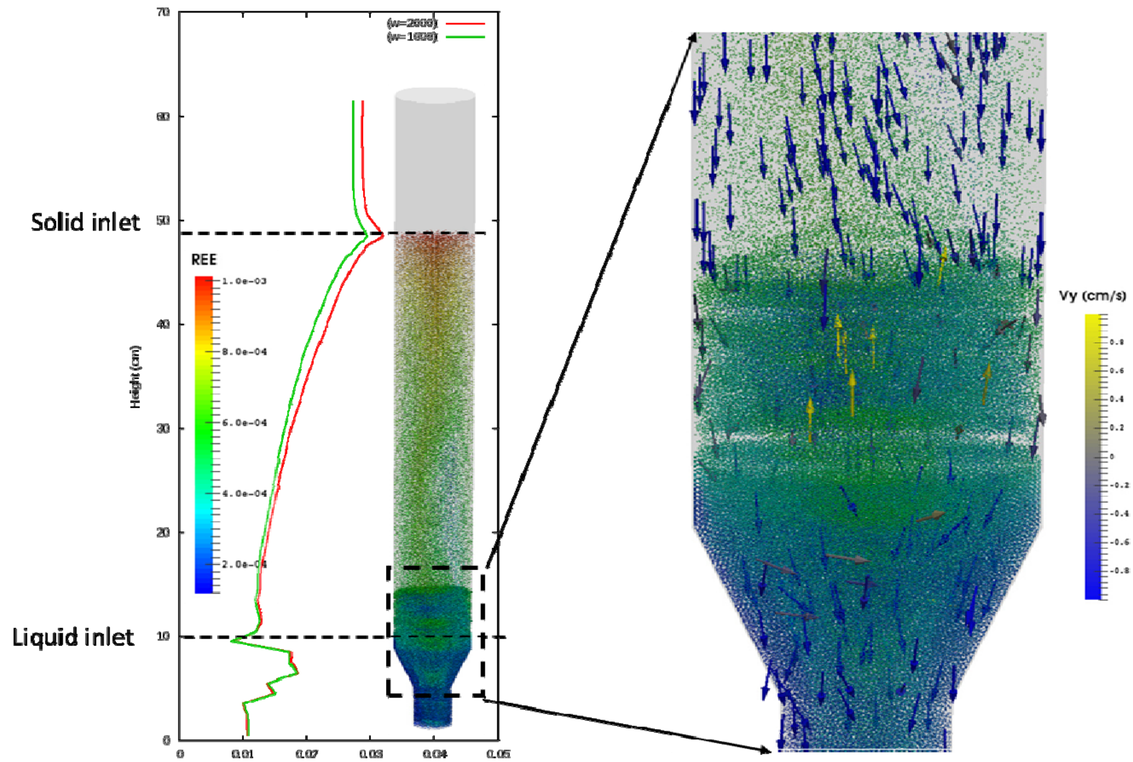
**Increasing REE concentrations**



Base
SIH58
L36
L36S36
W20
Solid inlet
Liquid inlet
REE
Y
X
Z

# *Counter-current fluidized bed case Base: solids distribution*



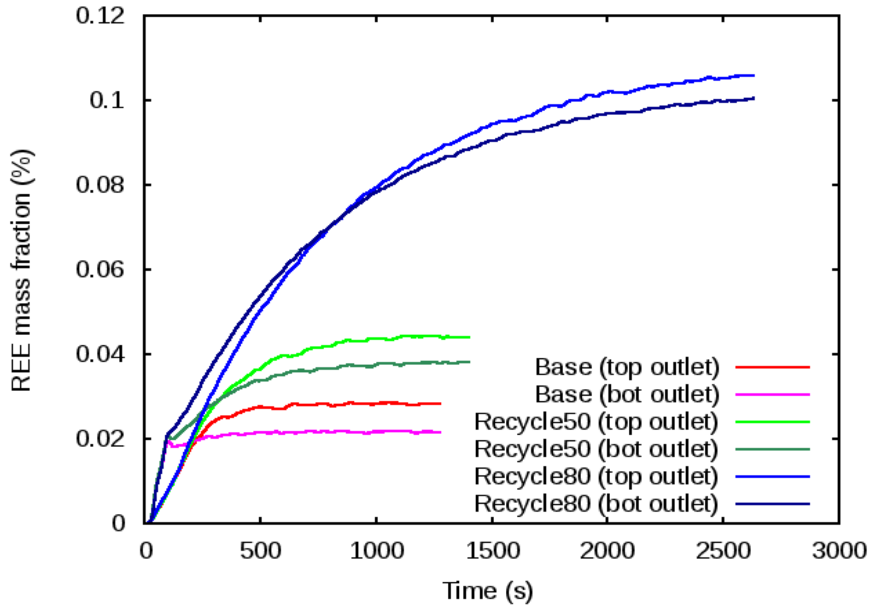




Time averaged REE mass fraction in liquid

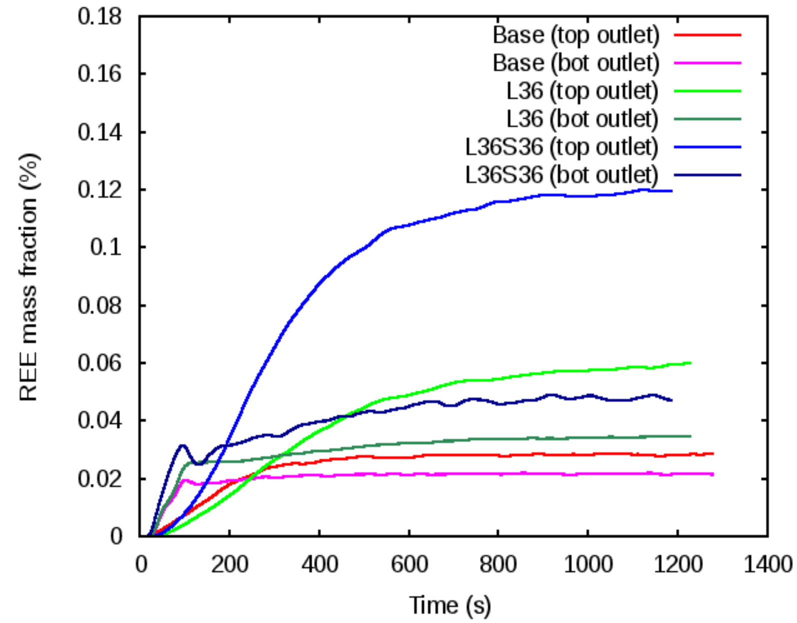
Instant REE mass fraction and particle velocity at t = 100 s

**There is a dense phase near the liquid inlet and above the dense phase is the dilute phase**

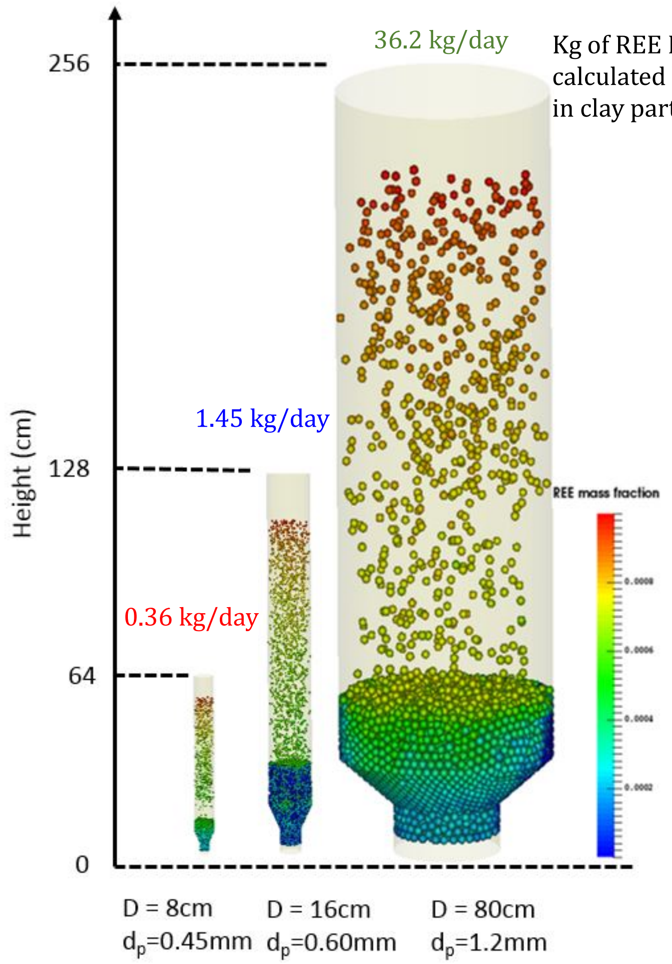
**The REE mass fraction gradient is well established in both dilute region and dense region**



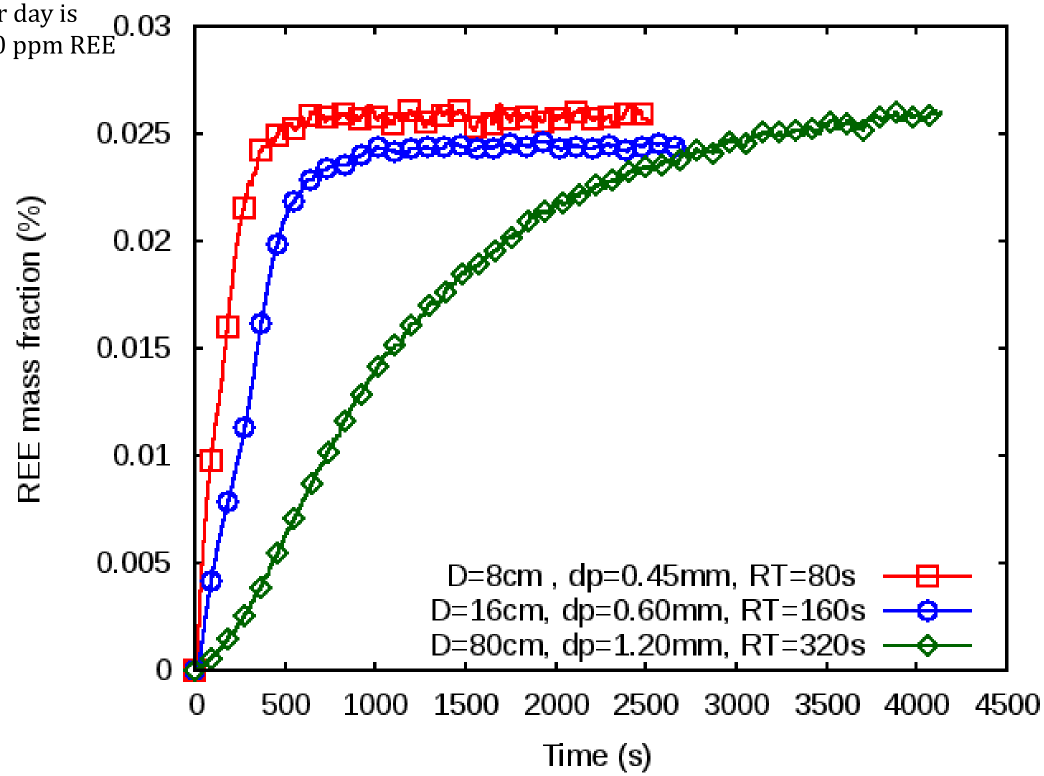

REE mass fraction at top outlet and bottom outlet with different liquid recycle fraction



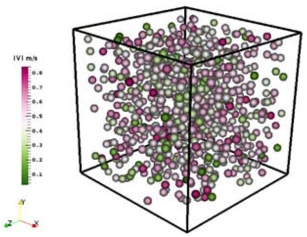

REE mass fraction with reduced liquid-solids ratio

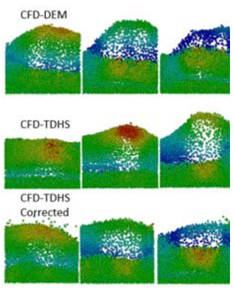


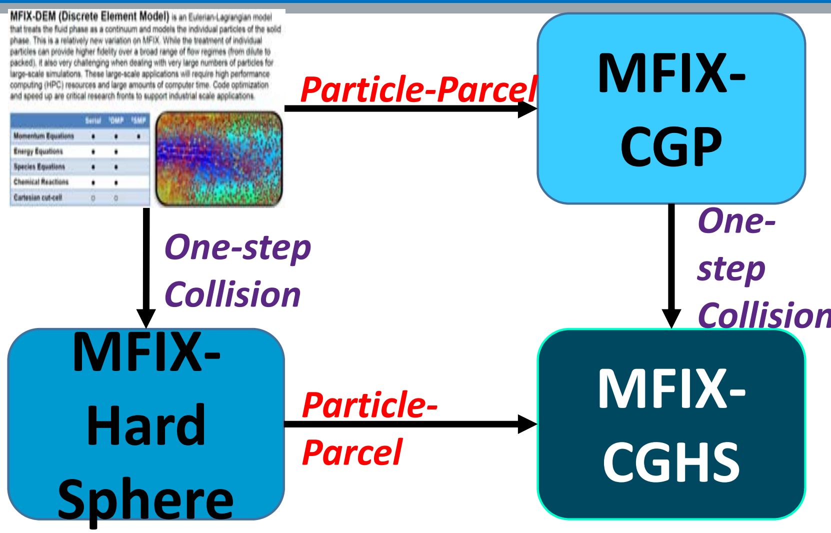

Kg of REE Production per day is calculated based on 1000 ppm REE in clay particles




- For scale-up, performances can be maintained with shorter reactor.
- If desired, larger particles may be used for larger reactor.



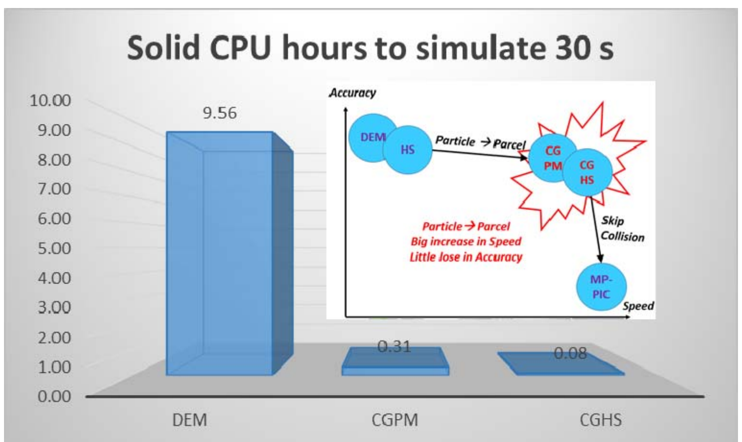


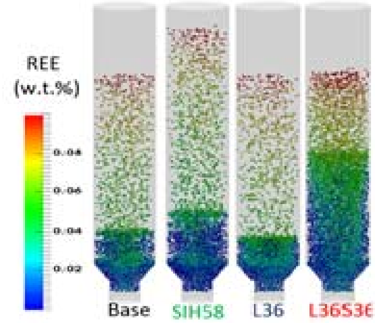



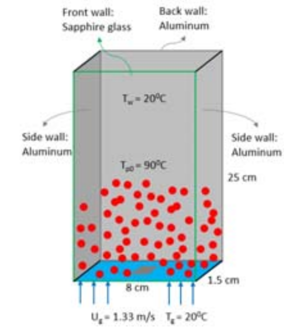
**MFIX-DEM (Discrete Element Model)** is an Eulerian-Lagrangian model that treats the fluid phase as a continuum and models the individual particles of the solid phase. This is a relatively new variation on MFIX. While the treatment of individual particles can provide higher fidelity over a broad range of flow regimes (from dilute to packed), it also very challenging when dealing with very large numbers of particles for large-scale simulations. These large-scale applications will require high performance computing (HPC) resources and large amounts of computer time. Code optimization and speed up are critical research fronts to support industrial scale applications.

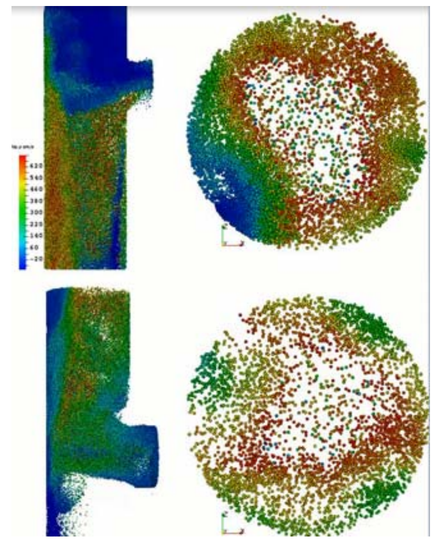
| | Serial | DMP | SMP |
|---|---|---|---|
| Momentum Equations | • | • | • |
| Energy Equations | • | • | |
| Species Equations | • | • | |
| Chemical Reactions | • | • | |
| Cartesian cut-cell | ○ | ○ | |



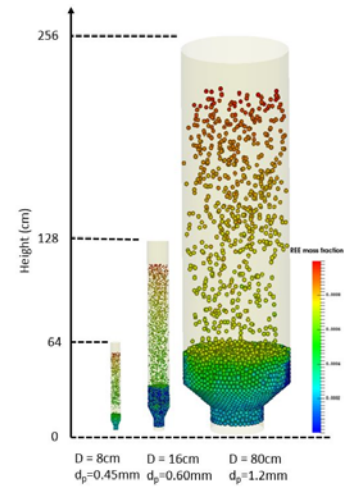


B22 riser




Scale-up REE reactor

## Accomplishments (peer-reviewed papers)

1. Lu, L; Yoo, K; Benyahia, S. Coarse grained particle method for simulation of liquid-solids reacting flows. *Ind. Eng. Chem. Res.* 2016, 55, 10477.

2. Lu, L; Li, T; Benyahia, S. An efficient and reliable predictive method for fluidized bed simulation. Submitted to *AIChE J.* (Jan. 2017).

3. Lu, L; Benyahia, S. Thousand-fold speedup of discrete-particle-based computer-aided reactor design and scale-up. Accepted at *Annual Tech Connect World Innovation Conference and Expo*. (Dec. 2016).

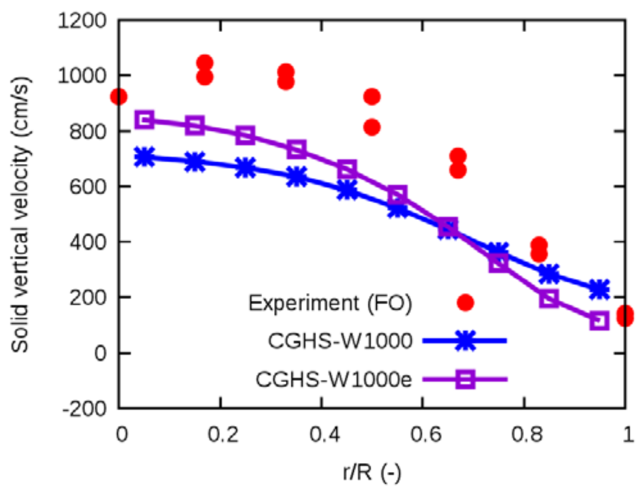
4. Lu, L; Konan, A; Benyahia, S. Influence of grid resolution, parcel size and drag models on bubbling fluidized bed simulation. Submitted to *Chem. Eng. J.* (Feb. 2017).

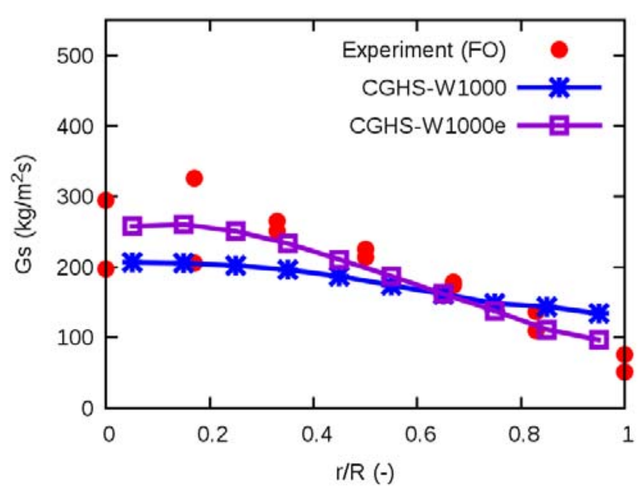
5. Lu, L; Morris, A; Li, T; Benyahia, S. Extension of a coarse-grained particle method to simulate heat transfer in fluidized beds. Submitted to *Int. J. Heat Mass Transfer* (Oct. 2016).

6. Lu, L; Gopalan, B; Benyahia, S. Assessment of different discrete particle methods ability to predict gas-particle flow in a small-scale fluidized bed. Submitted to *Chem. Eng. Sci.* (Mar. 2017).

# Any Questions?

# BACKUP SLIDES



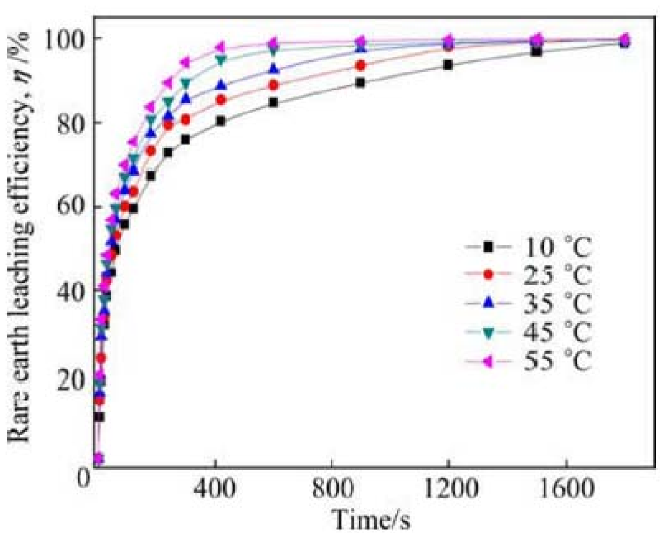




***Coarse-graining can yield accurate results with the ability to quantify uncertainties done in our previous work:***

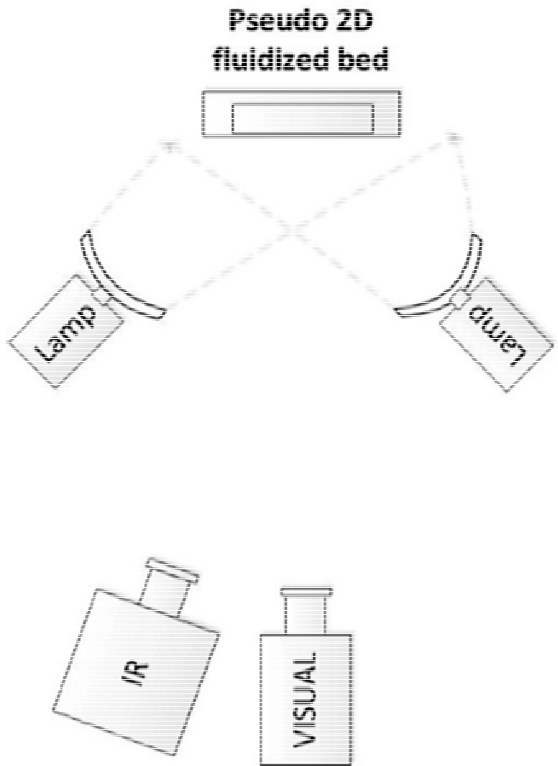
- Benyahia, Galvin: Ind. Eng. Chem. Res. 2010, 49, 10588–10605
- Lu, Xu, Ge, et al. Chem. Eng. Sci. 2016, 155, 314-337.

***Implementing Heat transfer in Coarse-grained model:***

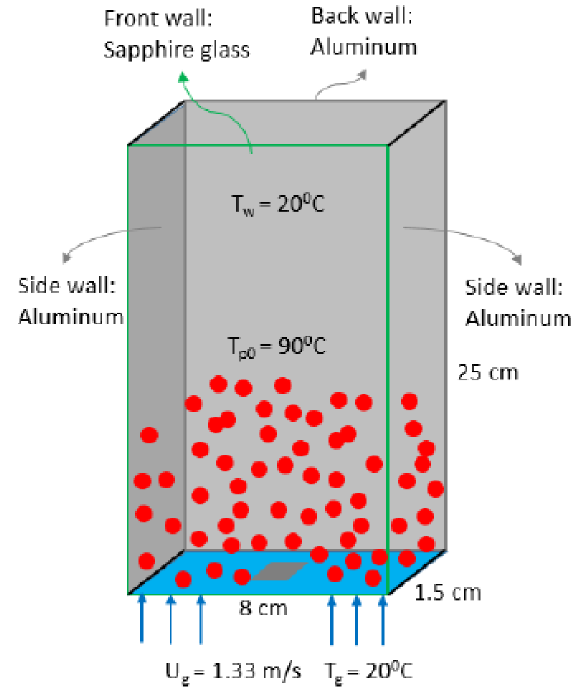
$$k_{exp}\left(1/\min\right) = \frac{0.011}{R_p\left(mm\right)^{1.217}} \exp\left(-\frac{9480}{RT}\right)$$




Xiao, Y.-f., Chen, Y.-y., Feng, Z.-y., Huang, X.-w., Huang, L., Long, Z.-q., Cui, D.-l., 2015. Leaching characteristics of ion-adsorption type rare earths ore with magnesium sulfate. Transactions of Nonferrous Metals Society of China 25, 3784-3790



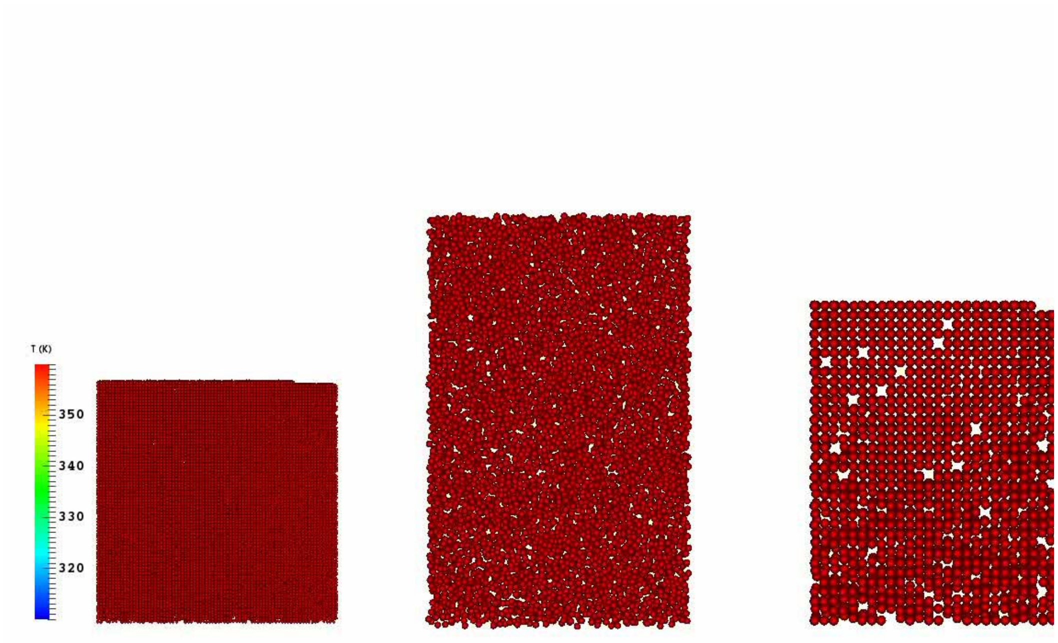

Experiment




Simulation

A.V. Patil, E.A.J.F. Peters, J.A.M. Kuipers, Comparison of CFD–DEM heat transfer simulations with infrared/visual measurements, Chem. Eng. J., 277 (2015) 388-401.

Constant Temperature wall boundary condition




CFD-DEM CGPM-8 CGPM-27

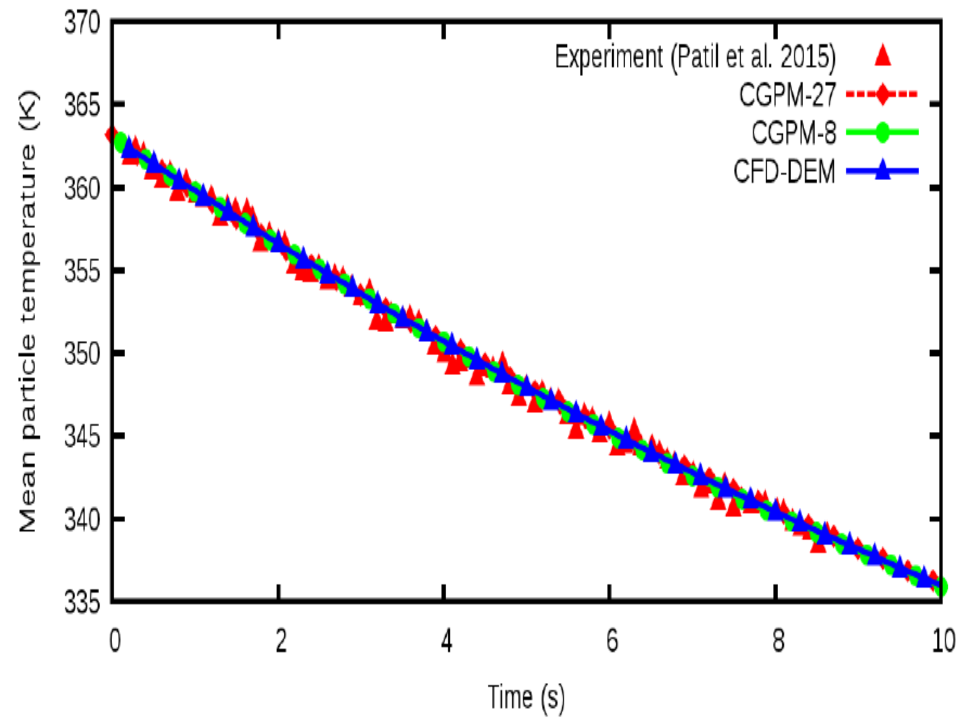
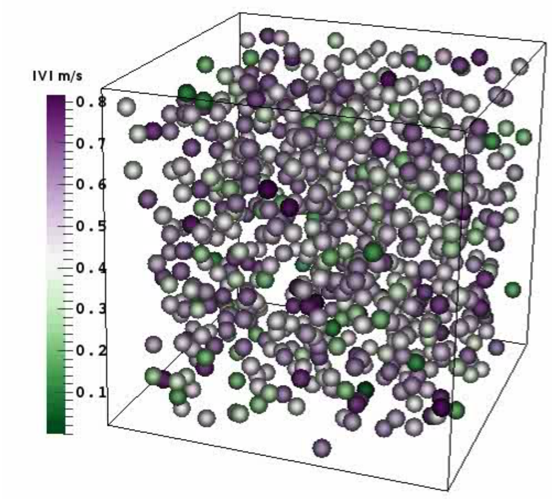
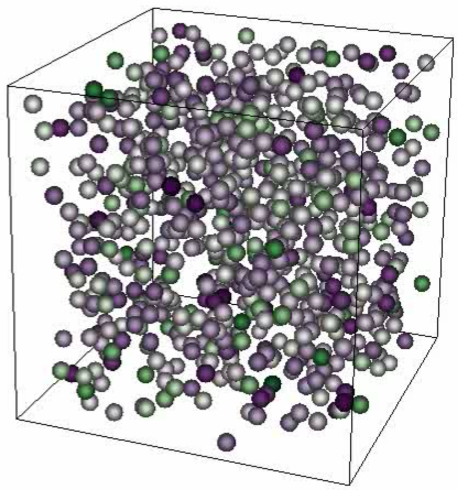



Fig. 8 Mean particle temperature profile simulated with different *W* and constant wall temperature boundary conditions

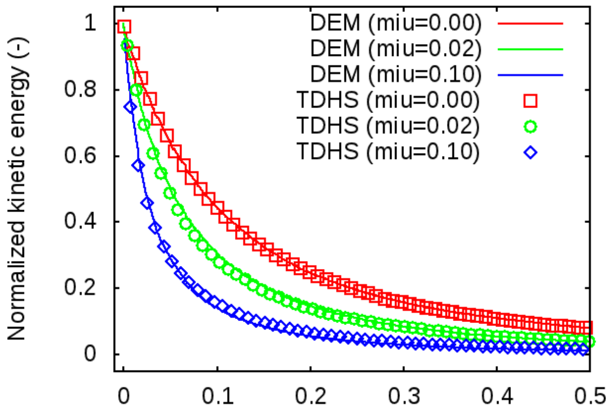
# *Hard sphere : Results*

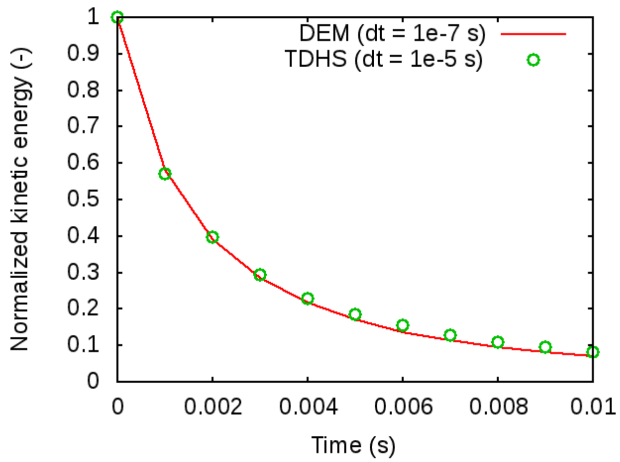



Soft-sphere

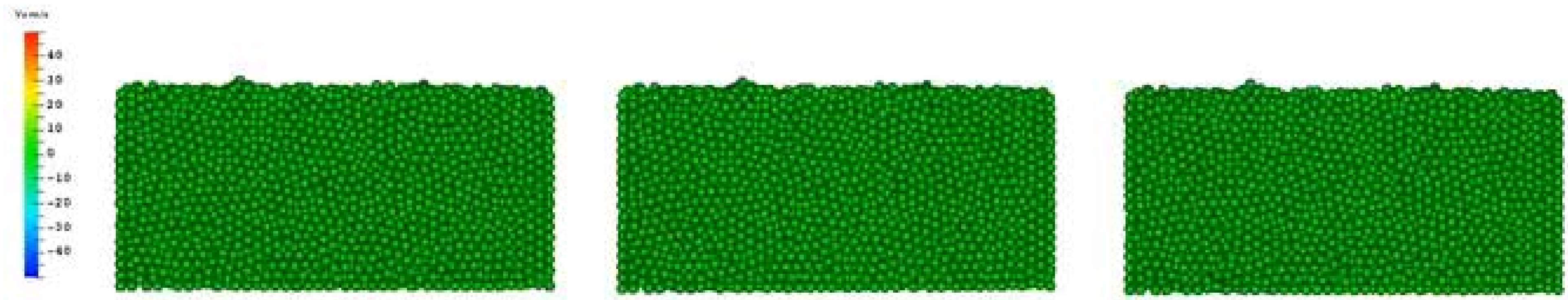


Hard-sphere




TDSH compute same results as DEM but with 100 times speedup!

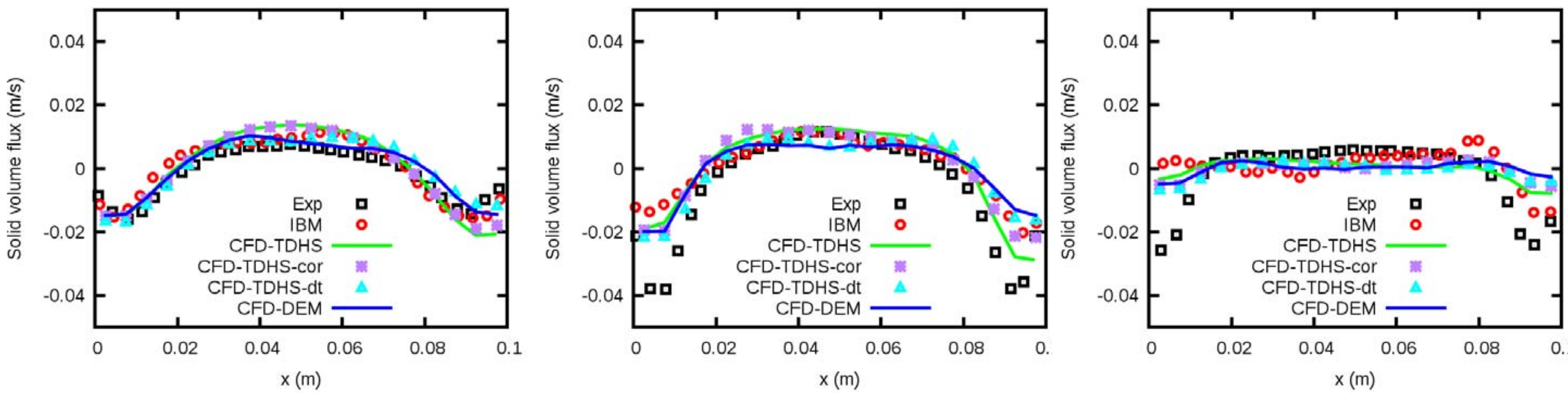



Soft-sphere
$\Delta t = 5 \times 10^{-6}$ s

Hard-sphere
$\Delta t = 1 \times 10^{-5}$ s

Hard-sphere
$\Delta t = 1 \times 10^{-4}$ s

## *Hard sphere : Results*




(a) Height = 0.025 m (b) Height = 0.055 m (c) Height = 0.085 m

The simulated results compared well with both experiment and DNS simulation results.