CFD Simulations of a Regenerative Process for Carbon Dioxide Capture in Advanced Gasification Based Power Plants

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Our team

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Program Objective

The overall objective of the program is to develop a Computational Fluid Dynamic (CFD) model and to perform CFD simulations to describe the heterogeneous gas-solid absorption and regeneration and WGS reactions in the context of multiphase CFD for a regenerative magnesium oxide-based (MgO-based) process for simultaneous removal of CO_2 and enhancement of H_2 production in coal gasification processes.

Scope of Work

The Project consists of the following four (4) tasks:

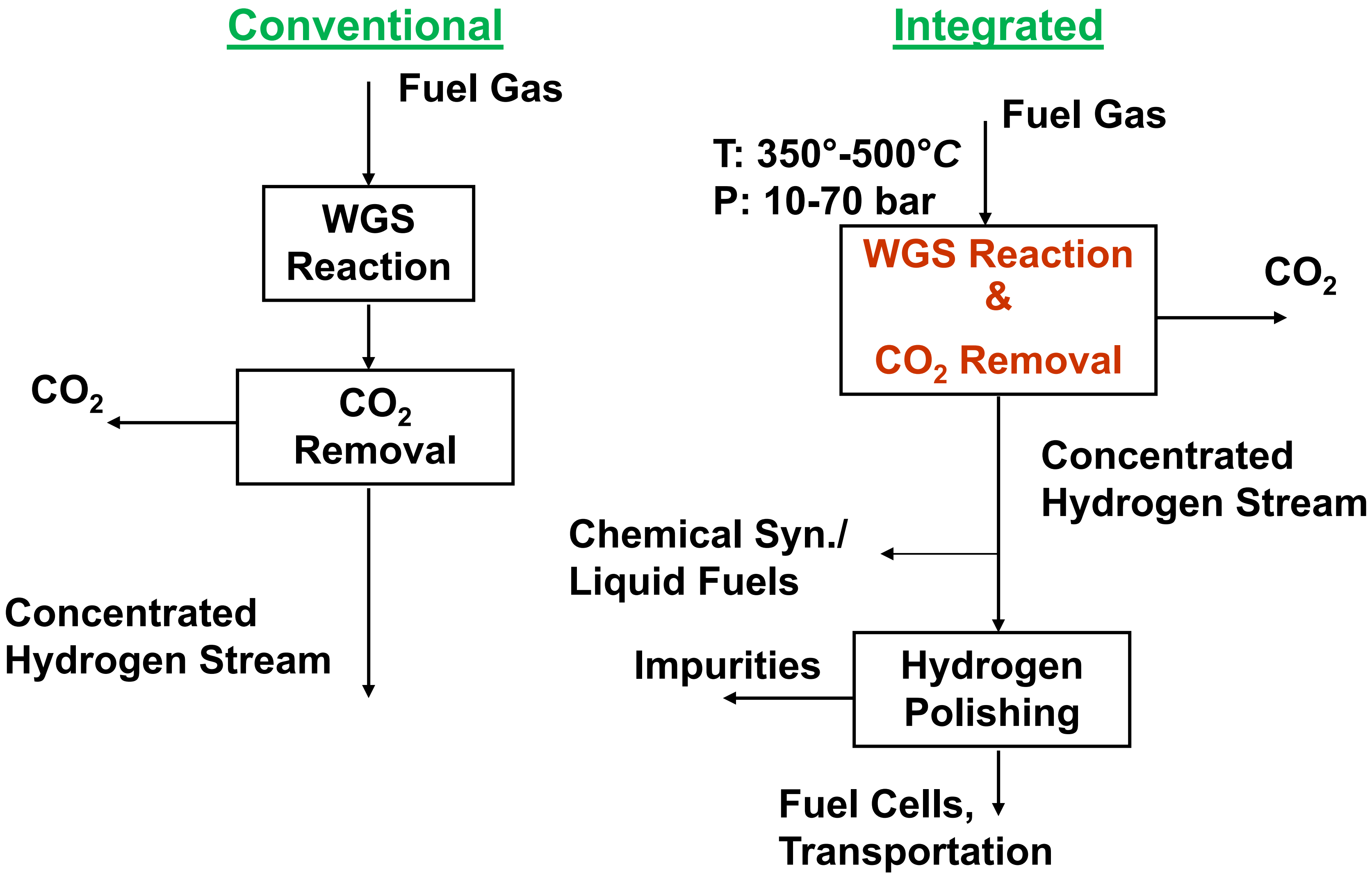
Task1. Development of a CFD/PBE model accounting for the particle (sorbent) porosity distribution and of a numerical technique to solve the CFD/PBE model.

Task2. Determination of the key parameters of the absorption and regeneration and WGS reactions.

Task3. CFD simulations of the regenerative carbon dioxide removal process.

Task4. Development of preliminary base case design for scale up

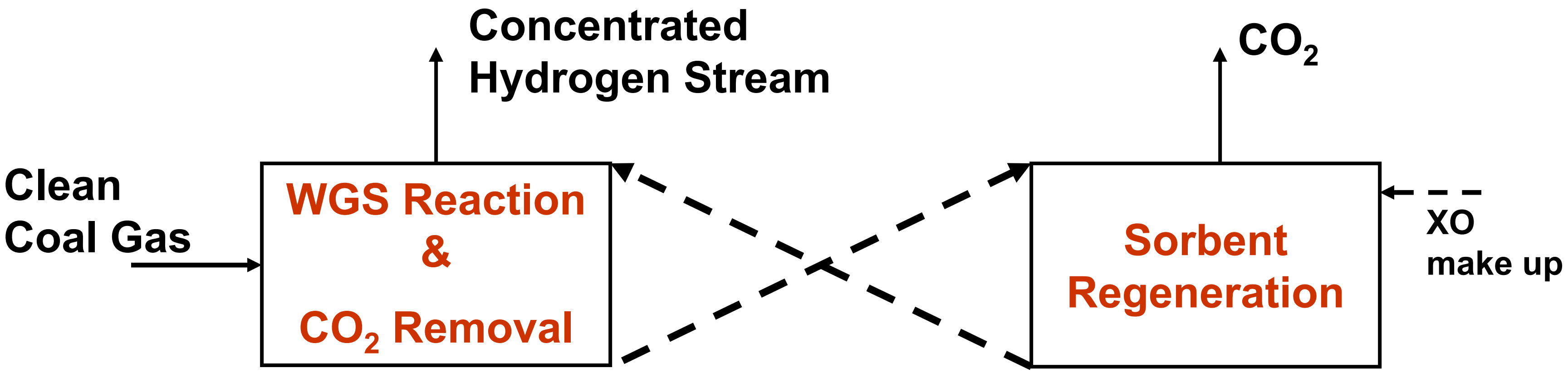
CO_2 Removal and Hydrogen Production



Conventional

Integrated

Regenerable Sorbent Approach



Clean Coal Gas

Concentrated Hydrogen Stream

WGS Reaction & CO_2 Removal

CO_2

Sorbent Regeneration

XO make up

$CO + H_2O \leftrightarrow CO_2 + H_2$

$XO + CO_2 \rightarrow XCO_3$

$XCO_3 \rightarrow XO + CO_2$

Part 1

Sorbent Preparation, Characteristics and Reactivity

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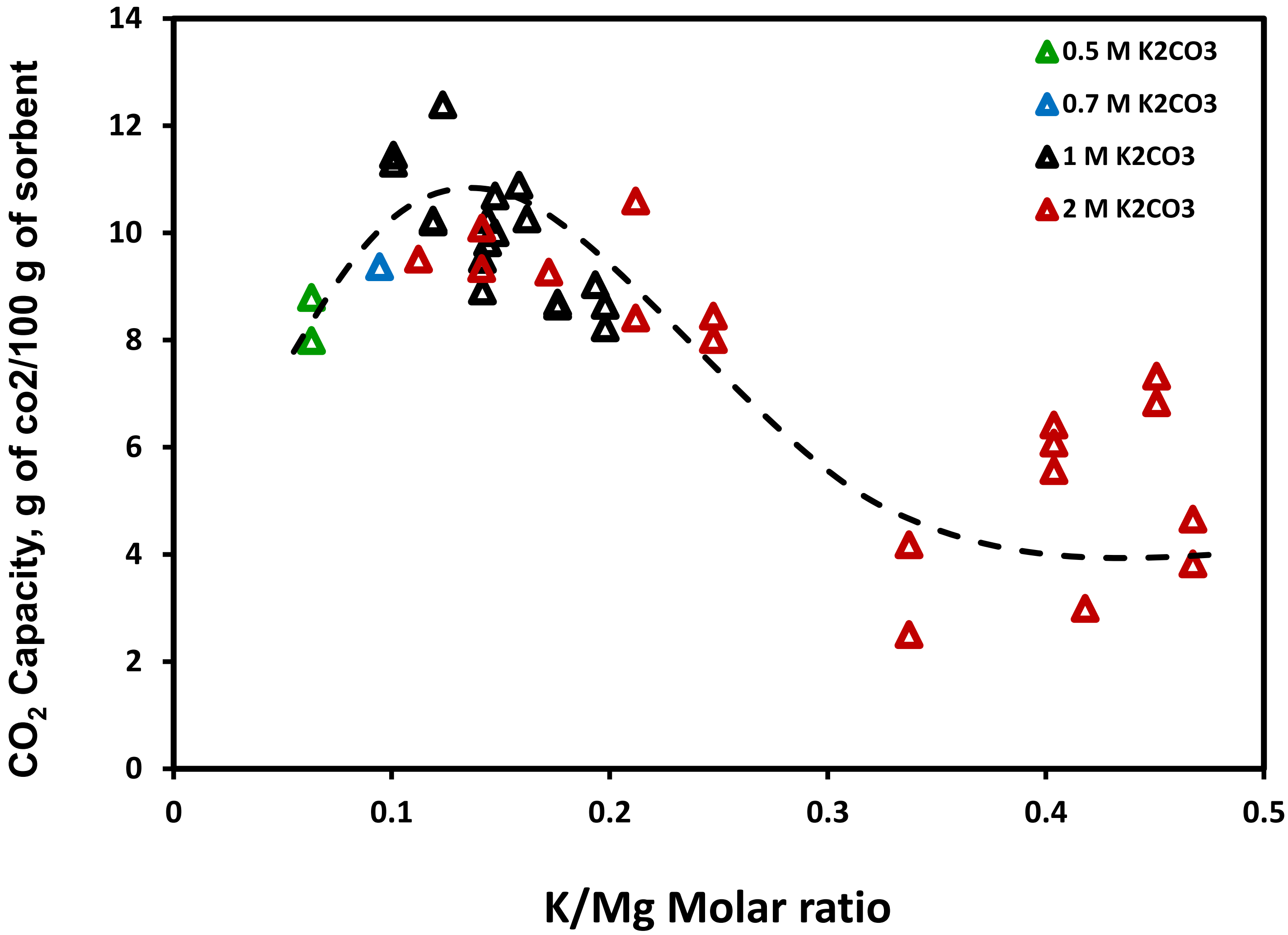
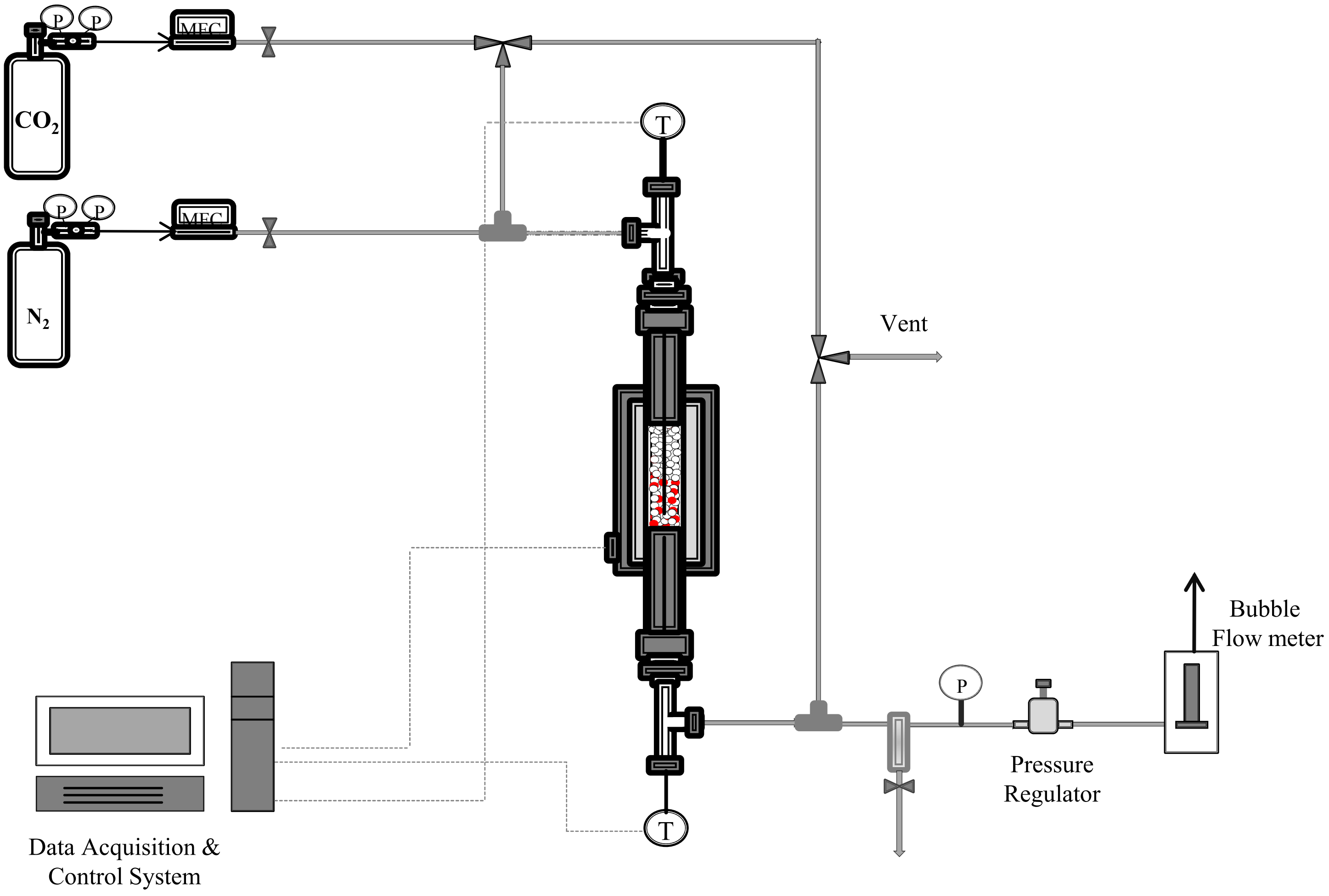
Effect of Potassium Concentration on Sorbent Capacity



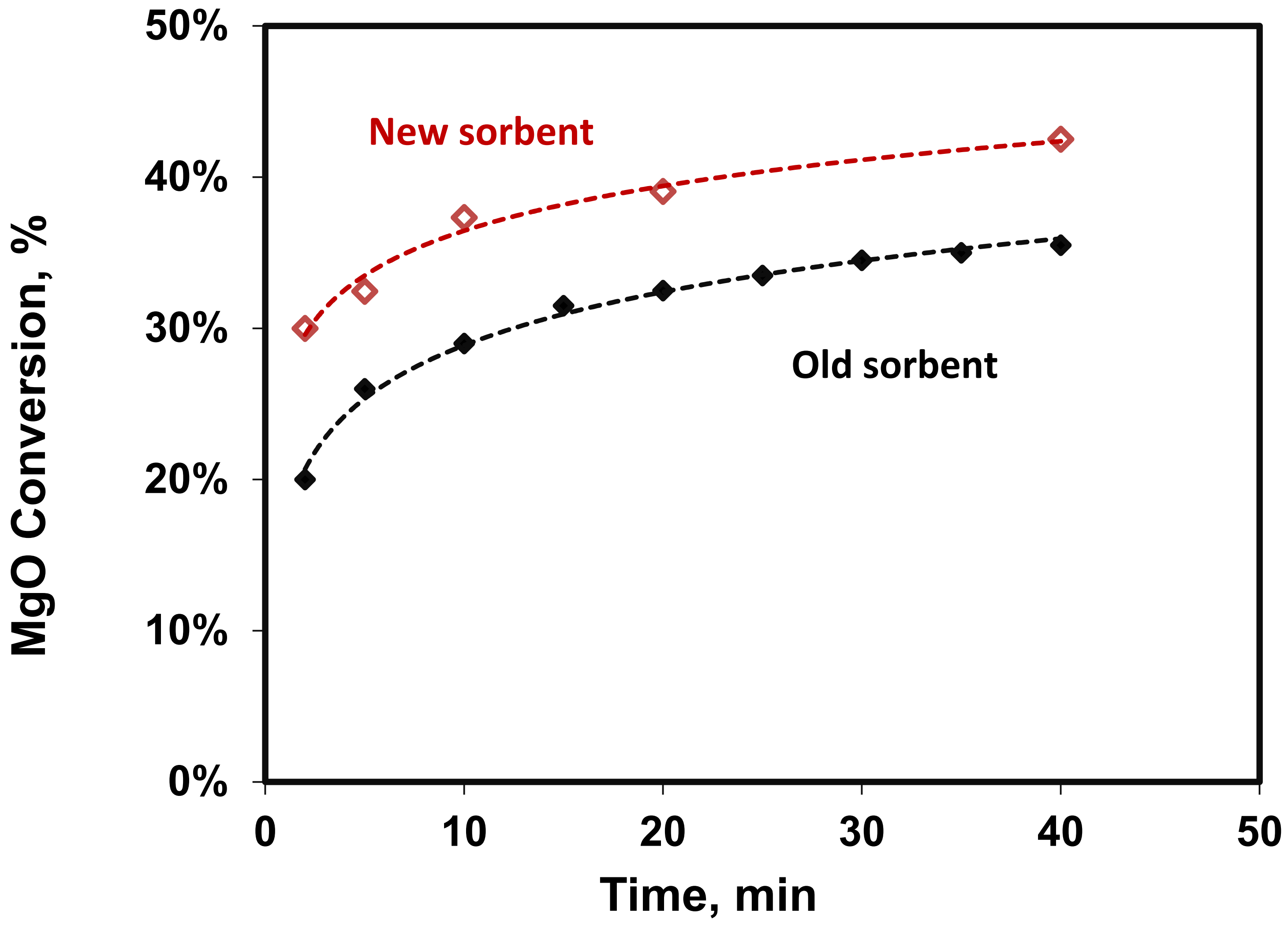
Optimum Sorbent Preparation Parameters

Preparation Parameters	HD52-P2
Sorbent particle diameter, μm	150-180
Calcination temperature, °C	520
Calcination temperature ramp, °C/min	1
Duration of calcination, hr	8
Concentration of potassium carbonate in the impregnation solution, mol/lit (M)	1
Duration of impregnation, hr	20
Drying temperature, °C (post-impregnation)	90
Humidity during drying, %	ambient
Duration of drying, hr	24
Re-calcination temperature, °C (post-drying)	470
Calcination temperature ramp, °C/min	1
Duration of re-calcination, hr	4

Experimental Setup: Dispersed Bed Reactor



Reactivity of the Sorbents (Old & New)



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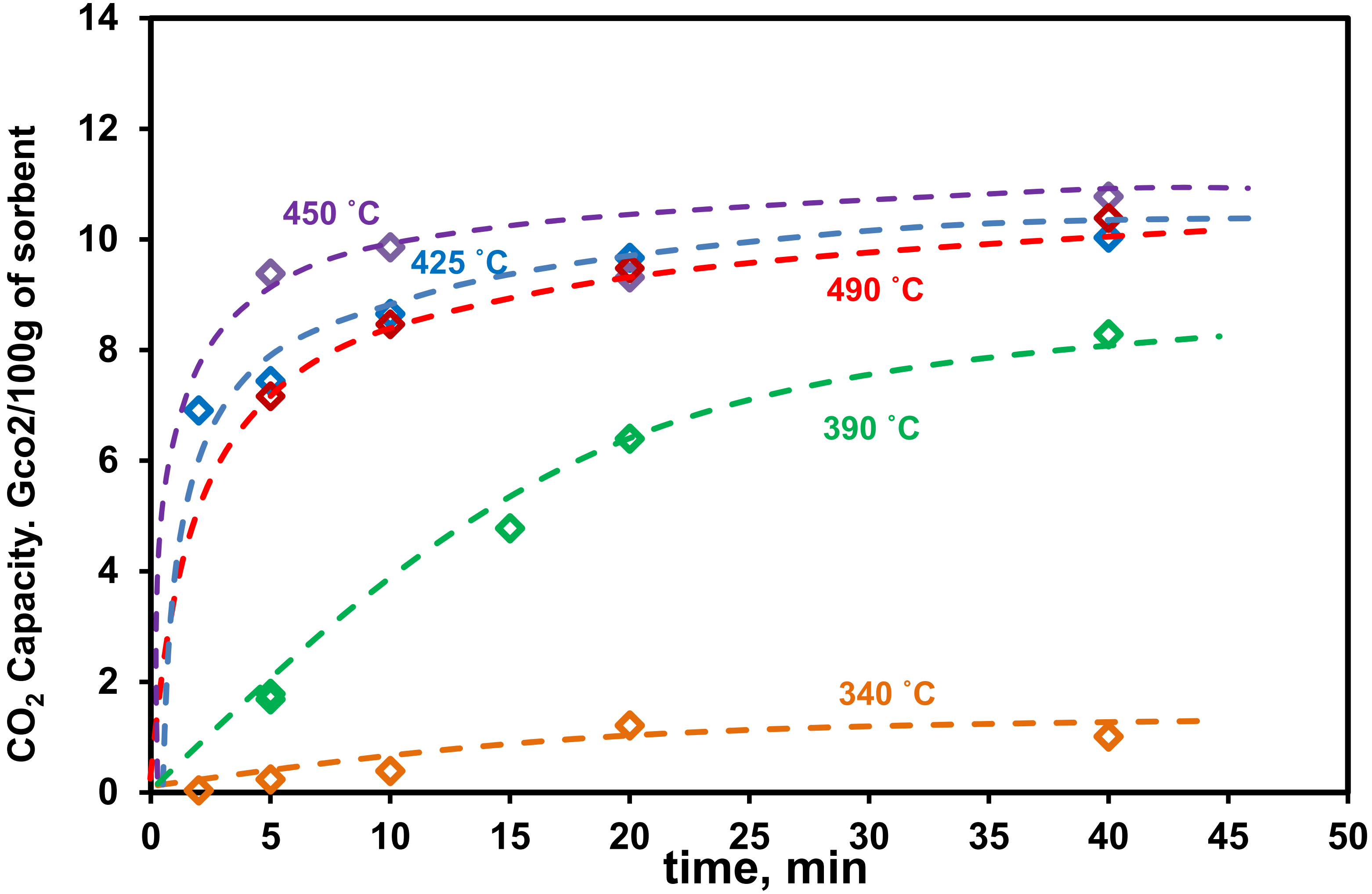
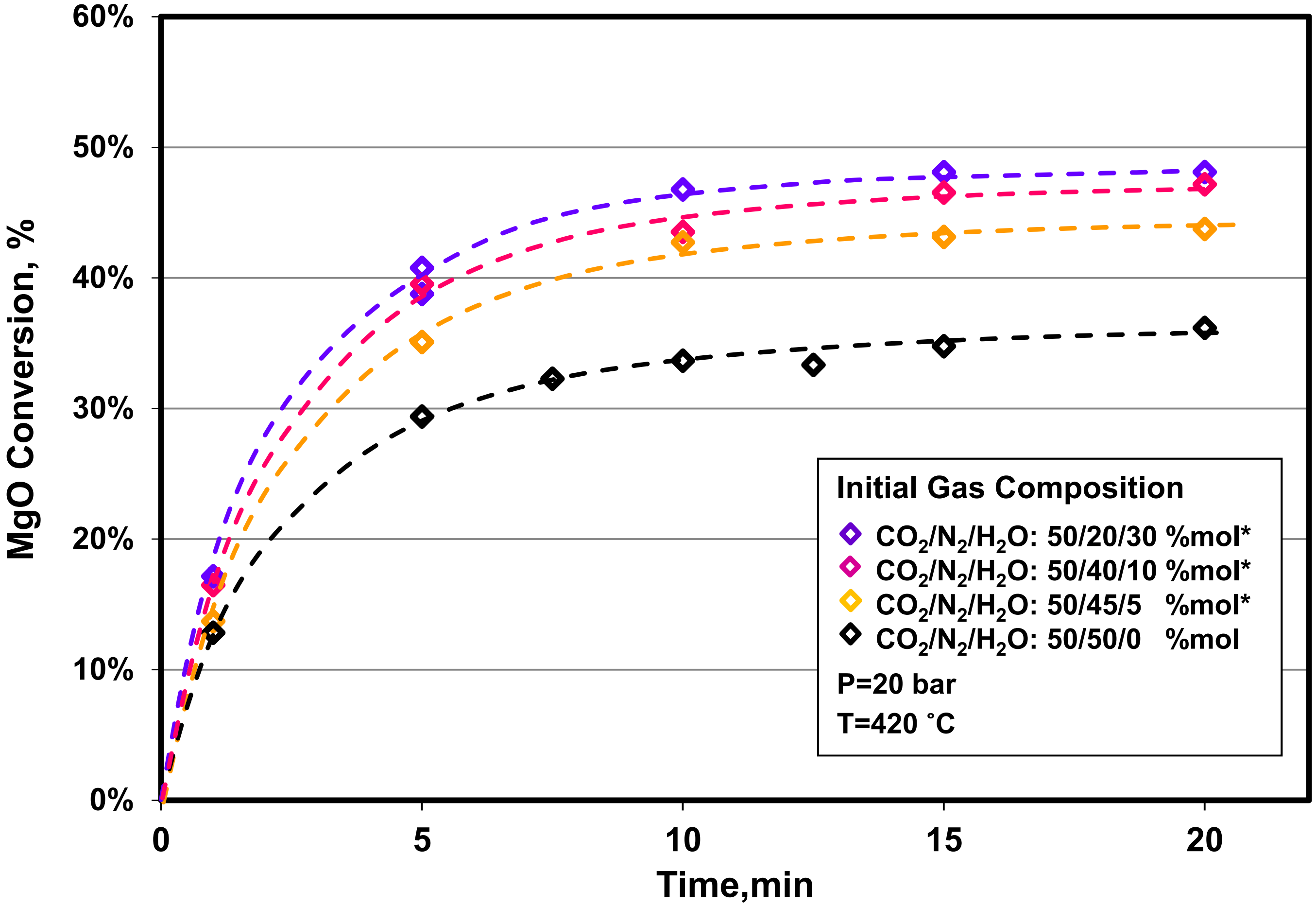
Effect of Temperature on Sorption



Effect of Steam on Reactivity

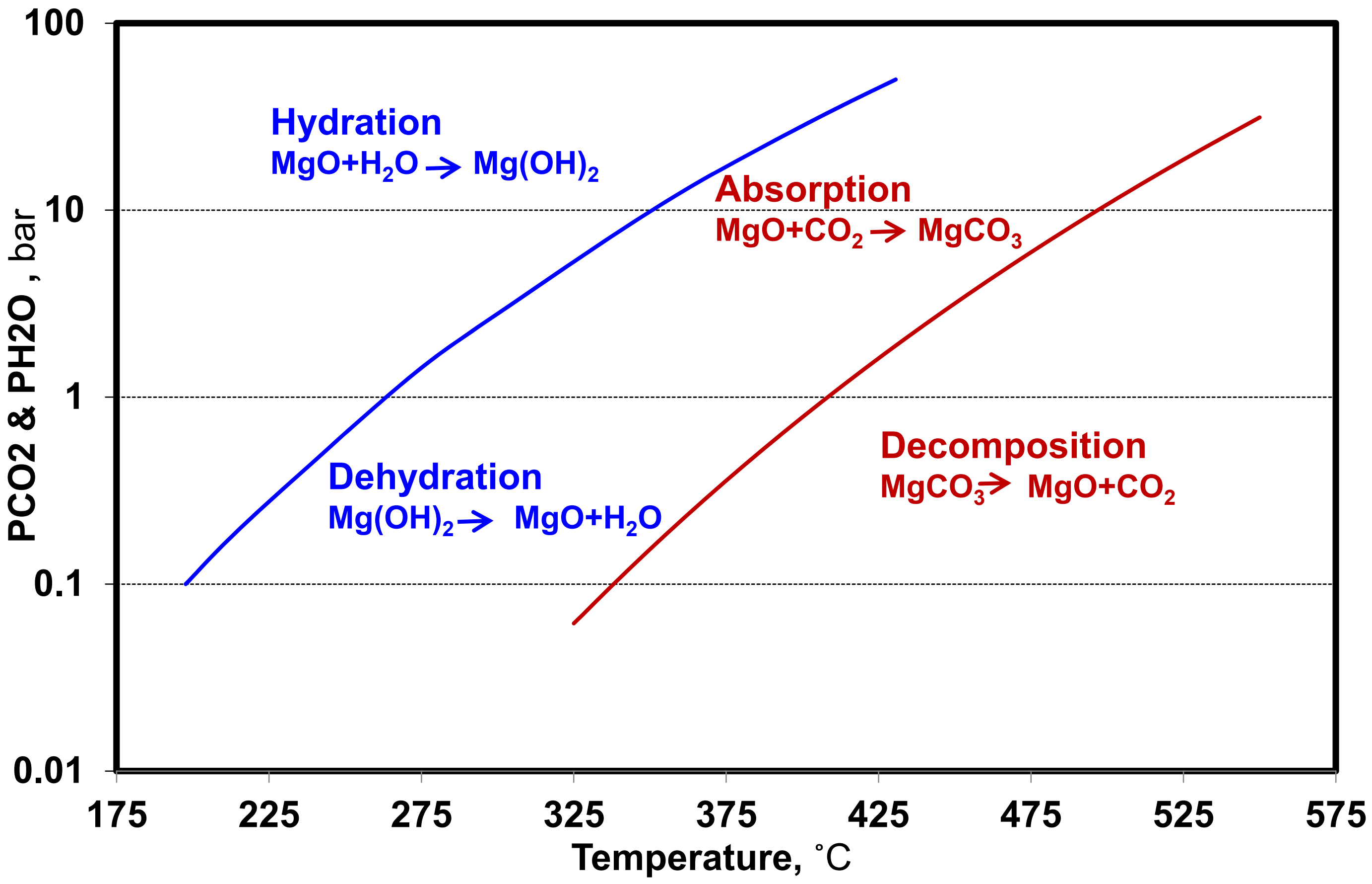


*The sorbent is exposed to steam for 30 min prior to the run.

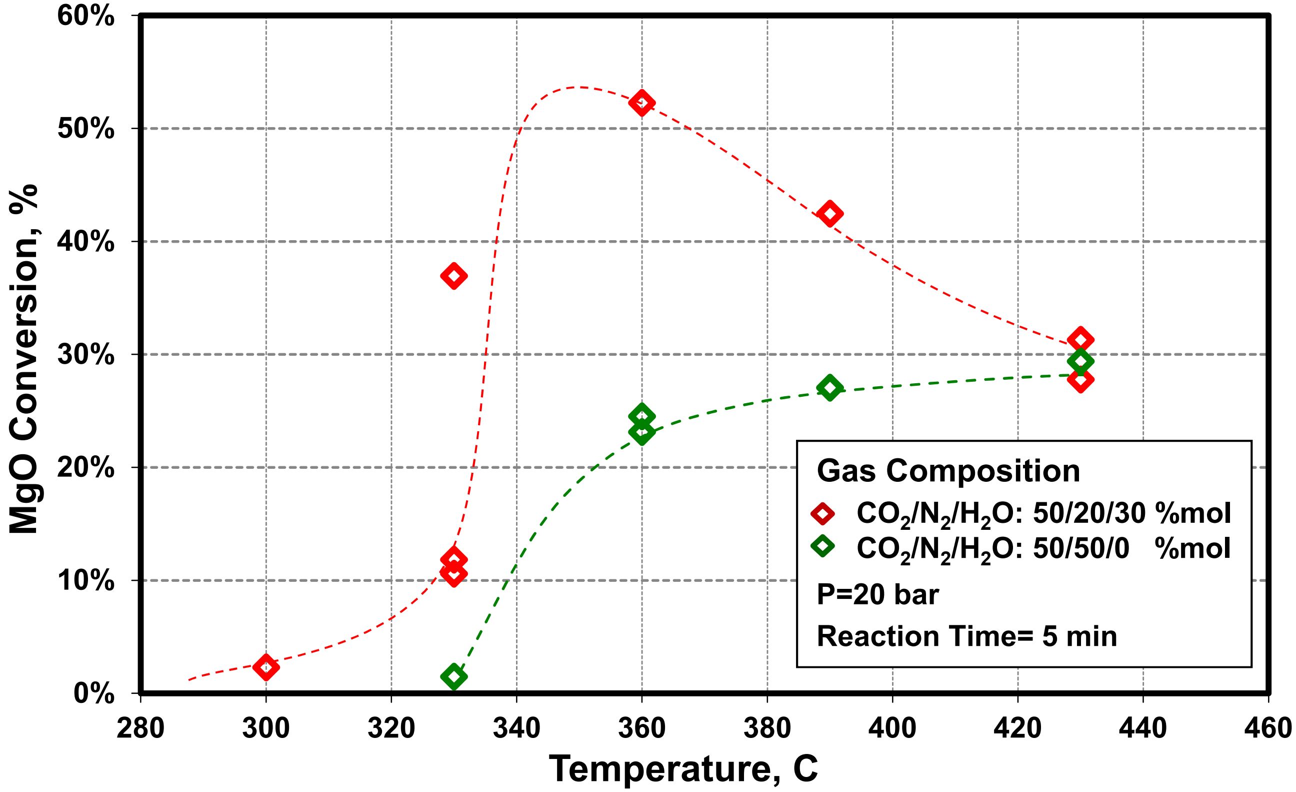
Possible Reasons for positive effect of steam

- Structural changes
- Secondary Carbonation Reaction
  - $MgO + H_2O = Mg(OH)_2$ Hydration
  - $Mg(OH)_2 + CO_2 = MgCO_3 + H_2O$ Carbonation

MgO-CO_2 & MgO-H_2O Equilibrium



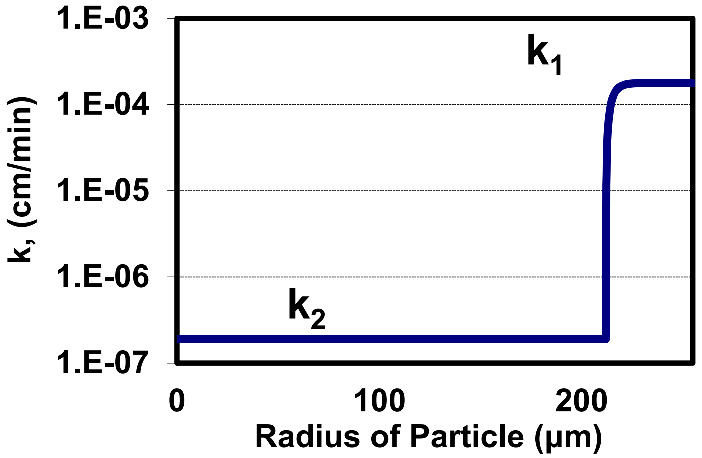
Effect of Steam on Sorbent Reactivity and Capacity

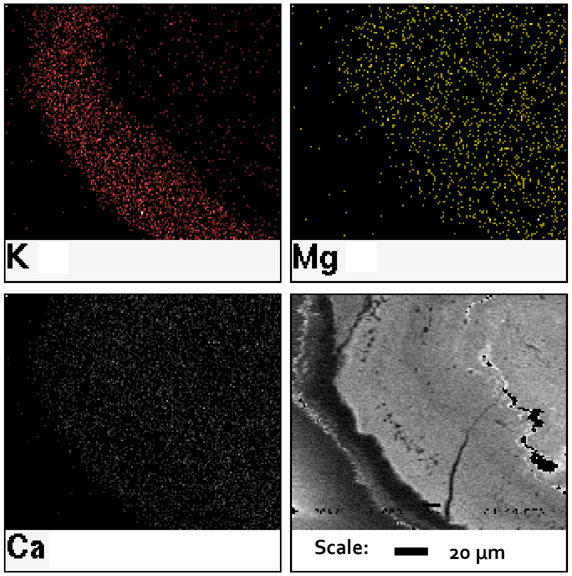


$MgO+CO_2 = MgCO_3$

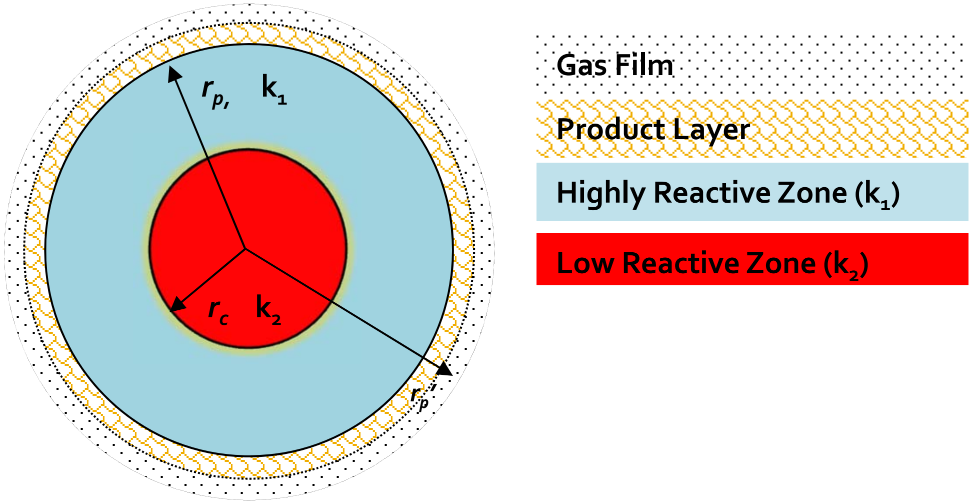
$Mg(OH)_2+CO_2 = MgCO_3 + H_2O$

Two-Zone Variable Diffusivity Shrinking Core Model with Expanding product layer





A. Hassanzadeh, 2007



$$D_g = D_{g0}(-\alpha X^{\beta}) \qquad r_p = r_p'\sqrt[3]{(1-X)+ZX}$$

$$Z = \frac{\rho_{product}\cdot M_{react}}{\rho_{react}\cdot M_{product}} \qquad k_s = \left|\begin{array}{ll} k_1 & for\ \ r \ge r_c \\ k_2 & for\ \ r < r_c \end{array}\right.$$

$$\boxed{\frac{dX}{dt} = -\frac{\dfrac{3}{r_p}\dfrac{k_s}{N^{o}_{MgO}}(C_b - C_e)(1-X)^{\frac{2}{3}}}{1+\dfrac{k_s}{D_g}r_p(1-X)^{\frac{1}{3}}\left(1-\sqrt[3]{\dfrac{1-X}{1-X+XZ}}\right)}}$$

Abbasi et al., Fuel, 2013

Part 2

Coupled
Computational Fluid Dynamics (CFD)
Population Balance Model (PBM)

(CFD-PBM)

Numerical Modeling: Conservation Equations

Eulerian- Eulerian Approach in combination with the kinetic theory of granular flow

Assumptions: Uniform and constant particle size and density

- Conservation of Mass

- gas phase: $$\frac{\partial}{\partial t}(\varepsilon_g \rho_g) + \nabla.(\varepsilon_g \rho_g v_g) = \dot{m}_g$$

- solid phase $$\frac{\partial}{\partial t}(\varepsilon_s \rho_s) + \nabla.(\varepsilon_s \rho_s v_s) = \dot{m}_s$$

- Conservation of Momentum

- gas phase: $$\frac{\partial}{\partial t}(\varepsilon_g \rho_g v_g) + \nabla.(\varepsilon_g \rho_g v_g v_g) = -\varepsilon_g \nabla P + \nabla.\tau_g + \varepsilon_g \rho_g g - \beta_{gs}(v_g - v_s)$$

- solid phase $$\frac{\partial}{\partial t}(\varepsilon_s \rho_s v_s) + \nabla.(\varepsilon_s \rho_s v_s v_s) = -\varepsilon_s \nabla P - \nabla P_s + \nabla.\tau_s + \varepsilon_s \rho_s g + \beta_{gs}(v_g - v_s)$$

- Conservation of Momentum

- gas phase: $$\frac{\partial}{\partial t}(\varepsilon_g \rho_g y_i) + \nabla.(\varepsilon_g \rho_g v_g y_i) = R_j$$

- solid phase $$\frac{\partial}{\partial t}(\varepsilon_s \rho_s y_i) + \nabla.(\varepsilon_s \rho_s v_s y_i) = R_j$$

- Conservation of solid phase fluctuating Energy

- solid phase $$\frac{3}{2}[\frac{\partial}{\partial t}(\varepsilon_s \rho_s \theta) + \nabla.(\varepsilon_s \rho_s \theta) v_s] = (-\nabla p_s I + \tau_s) : \nabla v_s + \nabla.(\kappa_s \nabla \theta) - \gamma_s$$

Generation of energy due to solid stress tensor

Diffusion

dissipation

Abbasi and Arastoopour , CFB10, 2011

Numerical Modeling: Drag Correlation

Gas-solid inter-phase exchange coefficient: EMMS model **(Wang *et al.* 2004)**

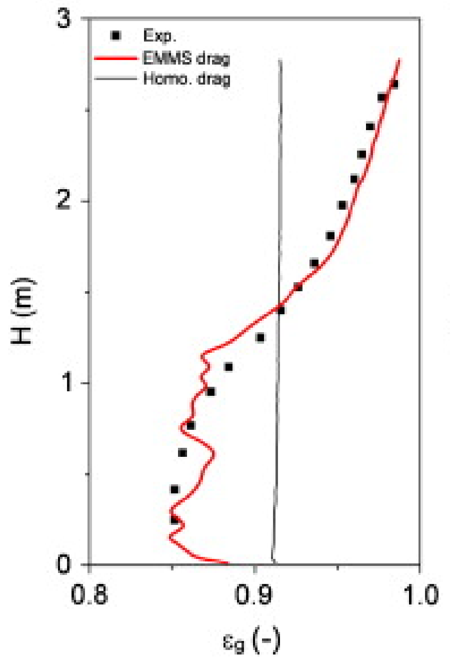
$$\beta_{sg} = \begin{cases} \dfrac{3}{4}\dfrac{(1-\varepsilon_g)\varepsilon_g}{d_p}\rho_g\left|u_g - u_s\right| C_{D0}\,\omega(\varepsilon_g) & \varepsilon_g > 0.74 \\ 150\dfrac{(1-\varepsilon_g)^2\mu_g}{\varepsilon_g d_p^{\,2}} + 1.75\dfrac{(1-\varepsilon_g)\rho_g\left|u_g - u_s\right|}{d_p} & \varepsilon_g < 0.74 \end{cases}$$

Heterogeneity Factor

ω < 1

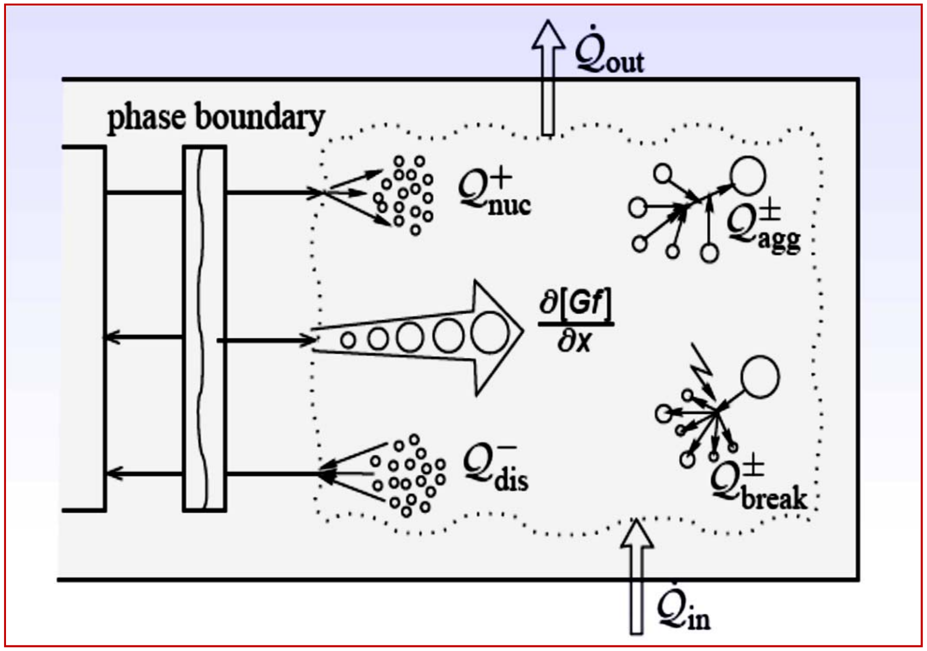
$$\omega(\varepsilon_g) = \begin{cases} -0.5760 + \dfrac{0.0214}{4(\varepsilon_g - 0.7463)^2 + 0.0044} & 0.74 < \varepsilon_g \le 0.82 \\ -0.0101 + \dfrac{0.0038}{4(\varepsilon_g - 0.7789)^2 + 0.0040} & 0.82 < \varepsilon_g \le 0.97 \\ -31.8295 + 32.8295\varepsilon_g & \varepsilon_g > 0.97 \end{cases}$$

Accounts for cluster formation by multiplying the "Wen & Yu" drag correlation with a heterogeneity factor



Li *et al.*, *Chem. Eng. Sci*, 2012

What is the Population Balance Equation?



To account for particle density distribution changes due to the reaction

$$\frac{\partial f(\xi;\mathbf{x},t)}{\partial t} + \frac{\partial}{\partial x_i}[u_p(t,\mathbf{x})f(\xi;\mathbf{x},t)] + \frac{\partial}{\partial x_i}[D_{pt}(\xi;\mathbf{x},t)\frac{\partial f(\xi;\mathbf{x},t)}{\partial x_i}] + \frac{\partial}{\partial \xi_j}[\frac{\partial \xi_j}{\partial t} f(\xi;\mathbf{x},t)] = h(\xi;\mathbf{x},t)$$

Accumulation term + Convection term + diffusive term + Growth term = Source term

Solution Method: FCMOM

Finite size domain Complete set of trial functions Method Of Moments: FCMOM

- Finite size domain: [-1, 1] instead of [0,∞]

$$\bar{\xi} = \frac{\{\xi - [\xi_{\min}(t) + \xi_{\max}(t)]/2\}}{[\xi_{\min}(t) + \xi_{\max}(t)]/2}$$

- Solution in terms of both Moments and size distribution
- $f(\xi,x,t)$ will be approximated by expansion based on a complete set of trial functions

$$f(\xi,x,t) = \sum_{n=0}^{\infty} C_n(t,x).\Phi_n(\xi) \qquad \text{when}$$

$$c_n = \sqrt{\frac{2n+1}{2}} \cdot \frac{1}{2^n} \cdot \sum_{v=0}^{n} (-1)^{n-v} \cdot \frac{(2v)!}{[(2v-n)!]} \cdot \left\{\frac{1}{[(n-v)!].[(v)!]}\right\} . \mu_{2v-n}$$

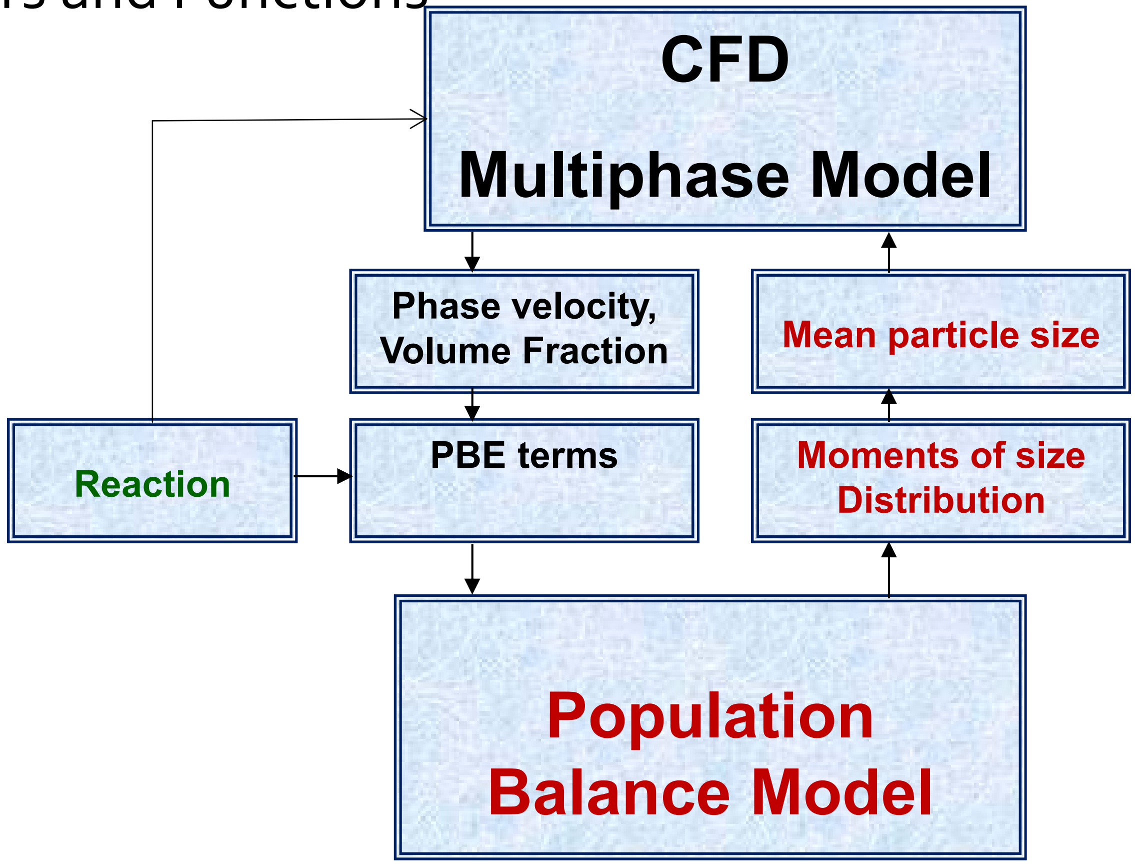
$$\mu_i = \int_{-1}^{1} \bar{f}'.(\bar{\xi})^i . d\bar{\xi} \qquad \phi_n(\bar{\xi}) = \sqrt{\frac{2n+1}{2}} . P_n(\bar{\xi})$$

$$\boxed{\frac{\partial \mu_i}{\partial t} + \nabla.(\mu_i . v_p) = -(MB + MB_{Conv} + IG)}$$

Strumendo and Arastoopour, 2008

Implementation and verification

- Implementation in Ansys /Fluent code via User Defined Scalars and Functions



$$d_{10} = \frac{m1}{m0}$$

$$\frac{\partial \varepsilon_s \rho_s \phi_s^i}{\partial t} + \nabla.(\varepsilon_s \rho_s v_p \phi_s^i - \varepsilon_s D_s^i \nabla \phi_s^i) = S_{\phi s}^i$$

Test case: Density Growth (Reaction) and convection

Assumption: Moments are convected with mixture velocity

$$\frac{\partial \mu_i}{\partial t} + \frac{\partial}{\partial x_j}[v_{p,j}\mu_i] = -\ \{[\overline{f'_{+1}} - (-1)^i.\overline{f'_{-1}}] - i.\mu_{i-1}\}.\frac{1}{(\xi_{\max} - \xi_{\min})}.(\frac{d\xi_{\min}}{dt}) -$$

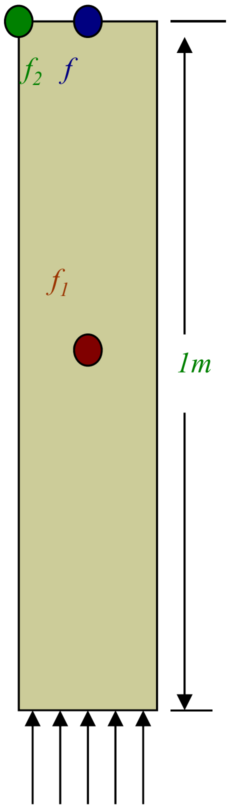
$$\{[\overline{f'_{+1}} - (-1)^{i+1}.\overline{f'_{-1}}] - (i+1).\mu_i\}.\frac{1}{(\xi_{\max} - \xi_{\min})}.(-\frac{d\xi_{\min}}{dt}) -$$

$$\{[\overline{f'_{+1}} - (-1)^i.\overline{f'_{-1}}] - i.\mu_{i-1}\}.\frac{v_{p,j}}{(\xi_{\max} - \xi_{\min})}.(\frac{\partial\xi_{\min}}{\partial x_j}) -$$

$$\{[\overline{f'_{+1}} - (-1)^{i+1}.\overline{f'_{-1}}] - (i+1).\mu_i\}.\frac{v_{p,j}}{(\xi_{\max} - \xi_{\min})}.(-\frac{\partial\xi_{\min}}{\partial x_j})$$

$$\frac{\partial\xi_{\min}}{\partial t} + v_p.\nabla\xi_{\min} = K$$

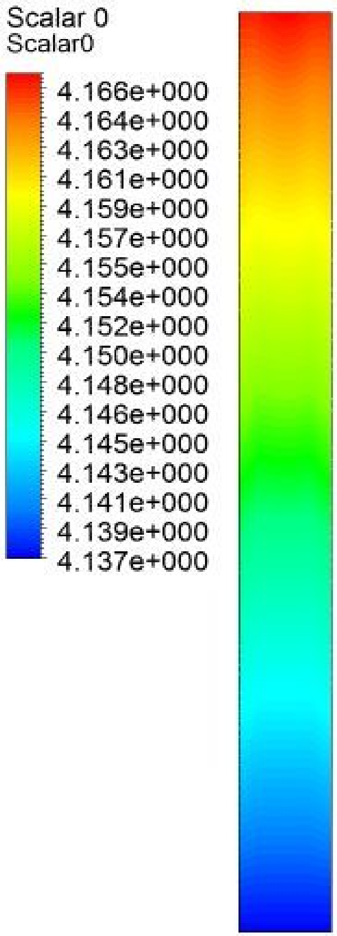
$$\rho_s = \frac{(\frac{\mu_1}{\mu_0})(\xi_{\max} - \xi_{\min}) + (\xi_{\min} + \xi_{\max})}{2}$$



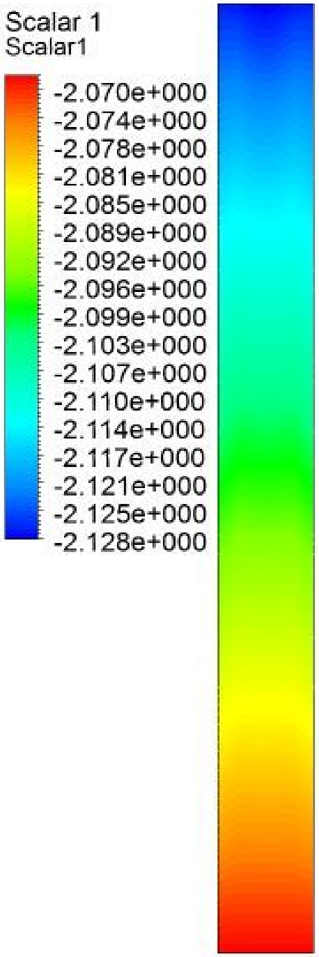
$v_g = v_s = 1$ m/s

$\varepsilon_s = 0.2$

Test case: Density Growth (Reaction) and convection

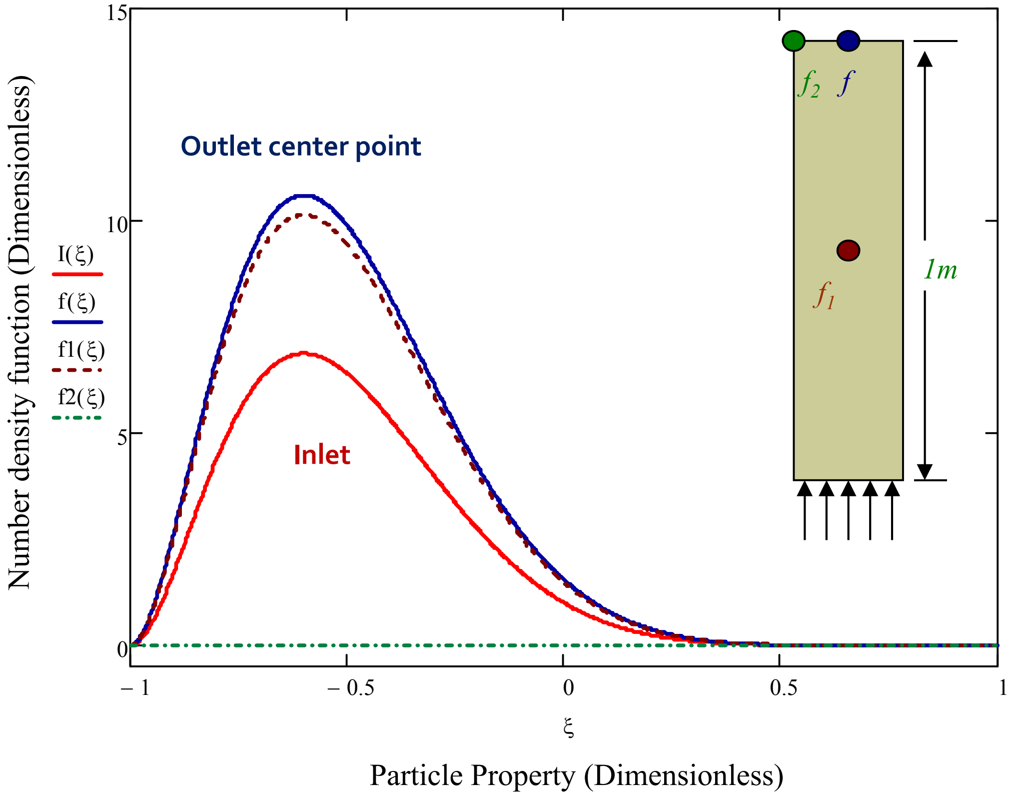


1st Moment



2nd Moment

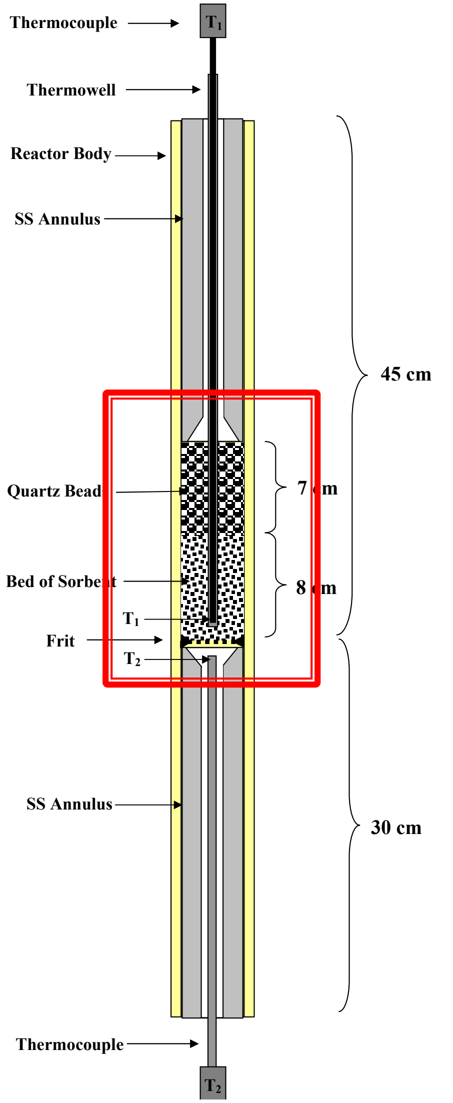
t = 10s

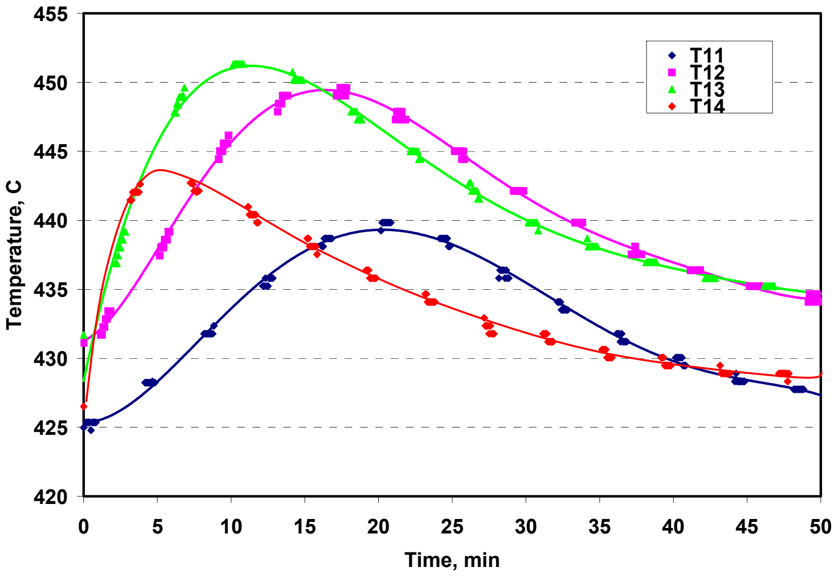


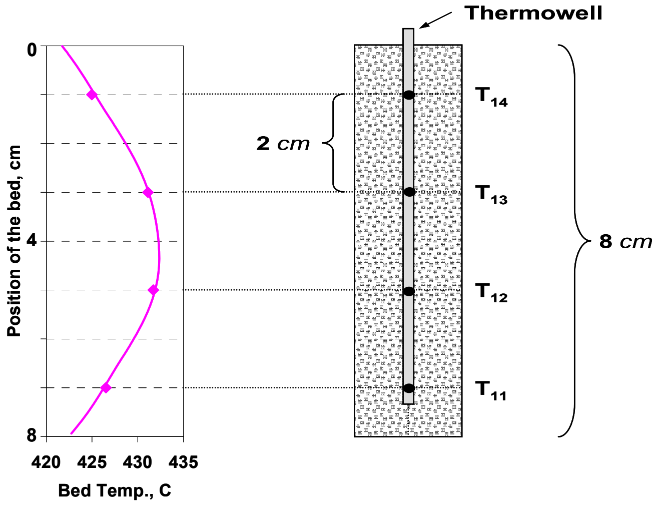
Part 3

Preliminary Base case design and Simulation Results

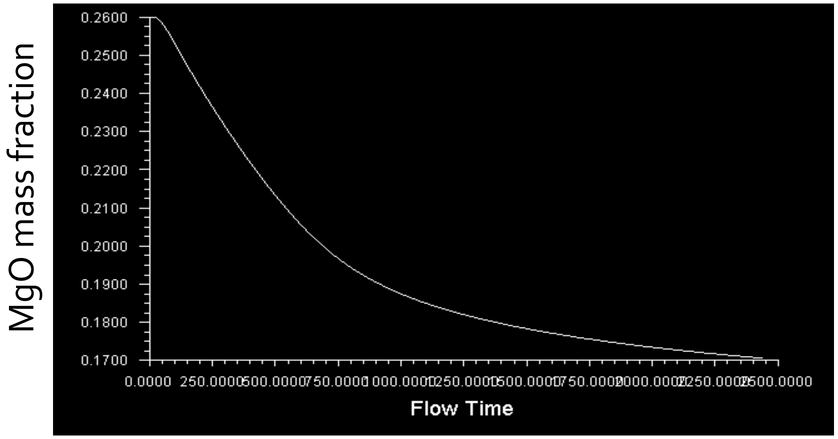
Packed-Bed Experiments and Modeling

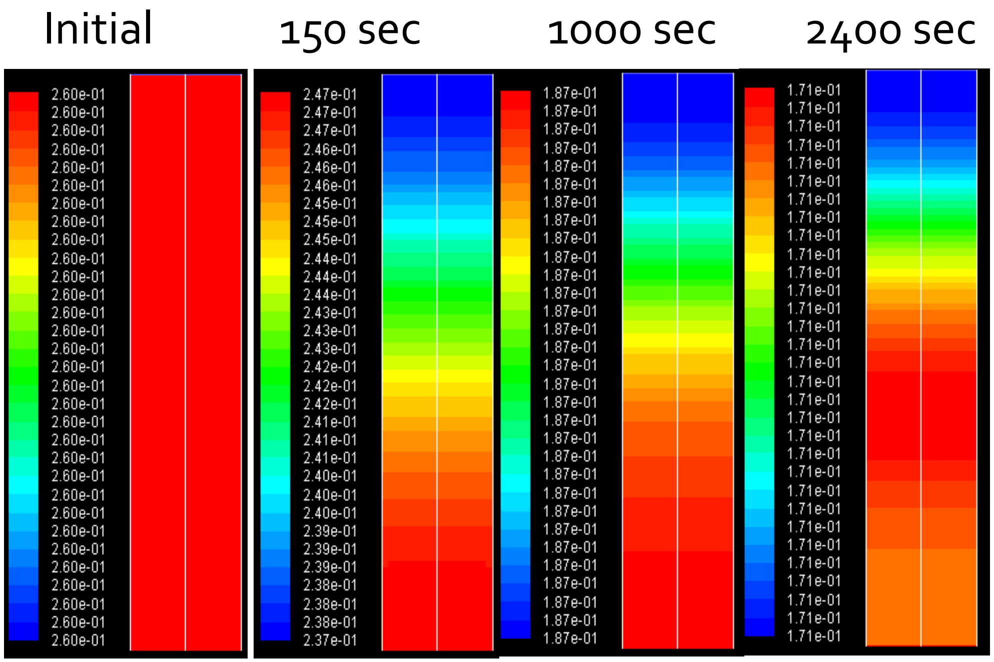




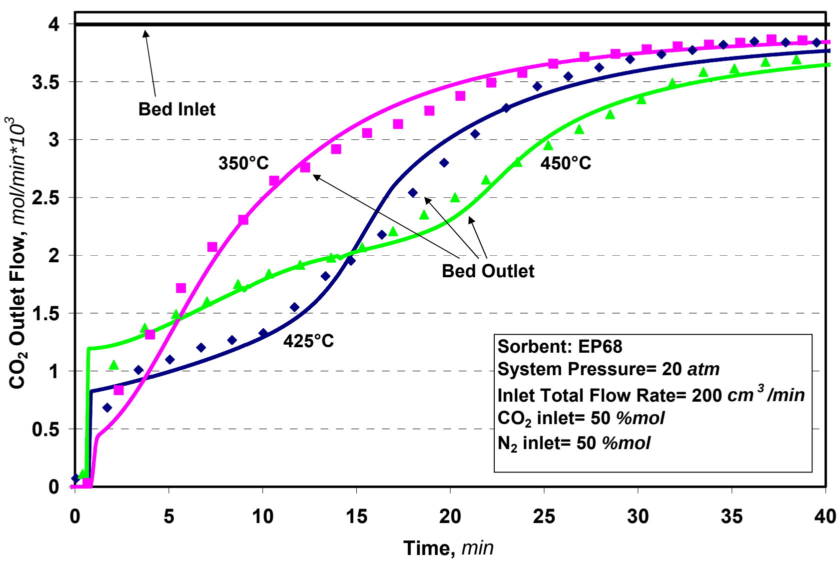


Packed-Bed Modeling results

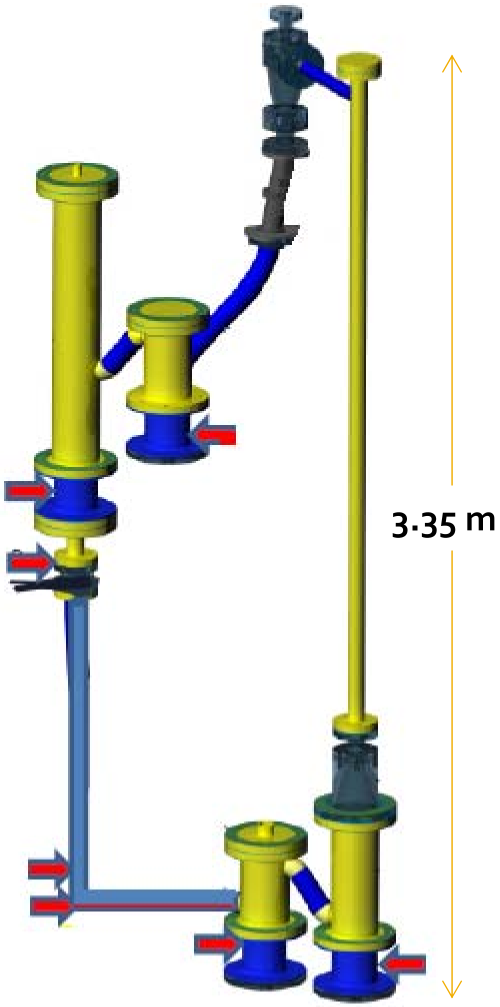


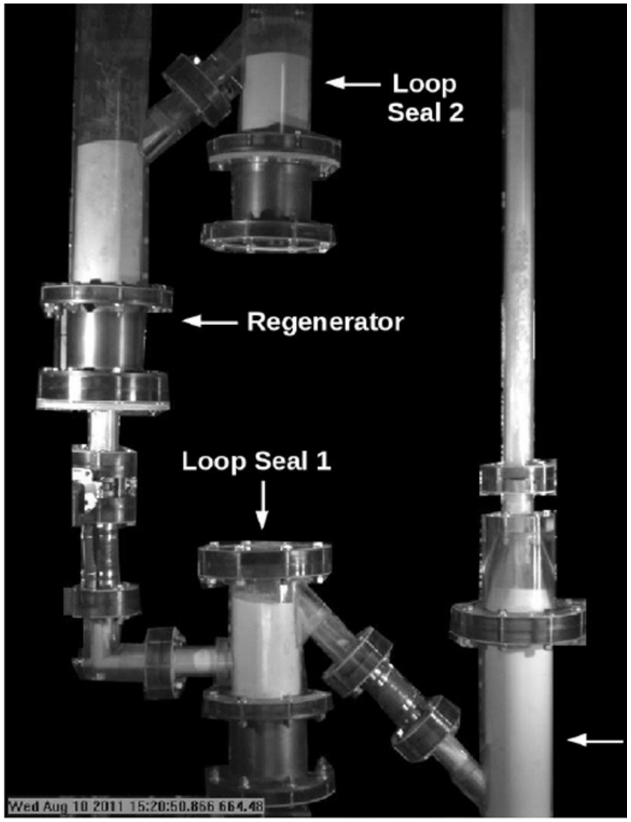


CO_2 Absorption Breakthrough Curve at Different Operating Temperatures



Full loop base case design



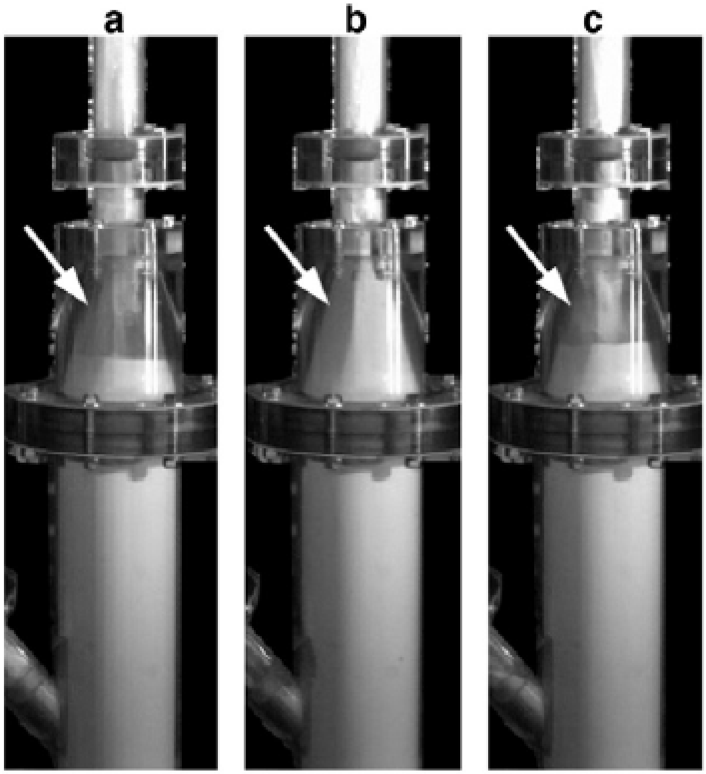


Location	Nominal Design gas Flow (g/s)
Adsorber	5
Loop seal 1	0.7
Loop seal 2	0.8
Regenerator	1
Move air	0.14

Based on DOE/ NETL Carbon Capture Unit.
(<u>Courtesy of Larry Shadle, NETL</u>)

Observed Fluidization behavior: Chugging

Sequence of events: (a) initially empty cone, (b) cone plugged with particles, (c) final empty cone.



NETL experimental images every 0.4-0.6 sec

"Chugging occurs when a large mass of particles lifts from the fluidized bed and moves into the cone leading into the riser. The cone-constriction prevents particles from flowing smoothly into the riser and particles plug the riser pipe."

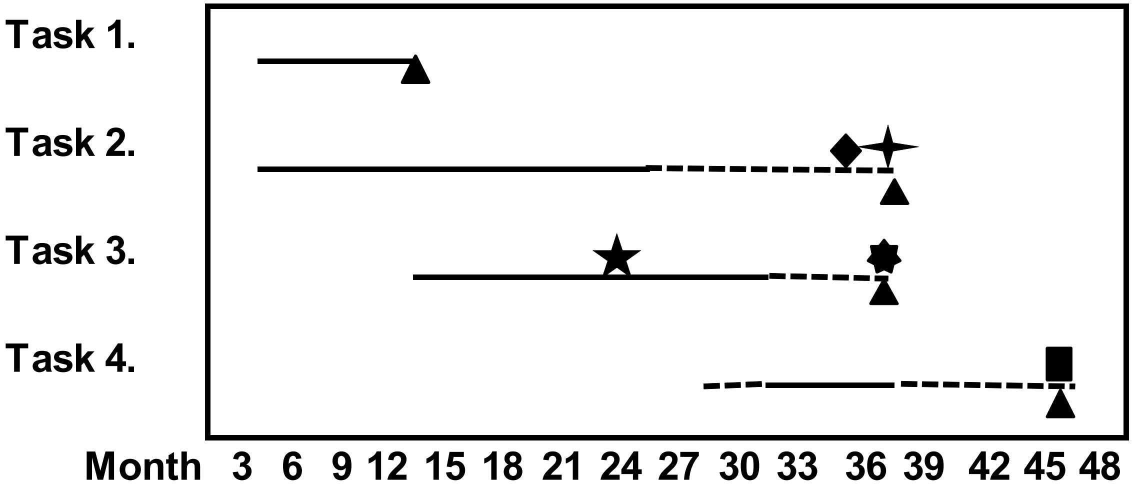
Clark et al., Powder Tech. 2013

Future Work

- **Modeling, simulation and base design**
  - Development of a modified frictional granular flow model and Completion of cold flow full loop CFB simulations for solid circulation rate calculations.
  - completion of riser simulation by including reaction and population balance model for density changes.
  - Development of preliminary base case design for scale up

- **Experiments**
  - Effect of CO2 and H2O concentration on absorption reaction and operating condition on regeneration reaction
  - Modeling of regeneration process and combined absorption & WGS reactions

Project Schedule



Milestones:

- ▲ Task completion
- ◆ Experimental work completed
- ✦ Reaction model finalized
- ★ CFD simulation of single reaction/reactor Completed
- ✸ CFD simulation of integrated process Completed
- ■ Development of the base-case design completed