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

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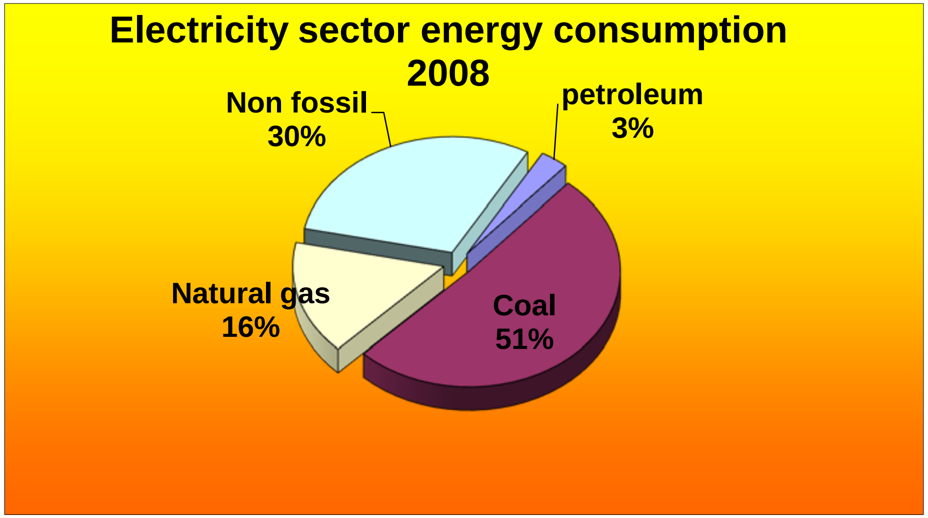
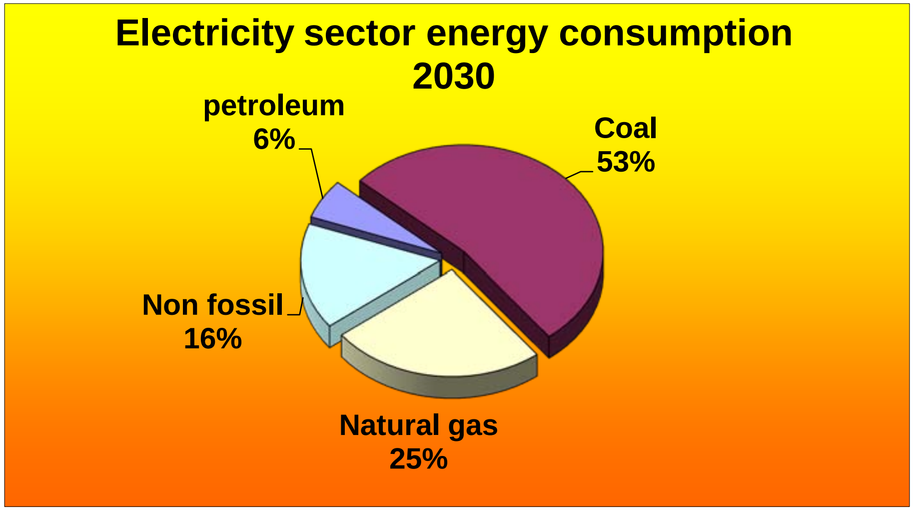
Javad Abbasian and Hamid Arastoopour

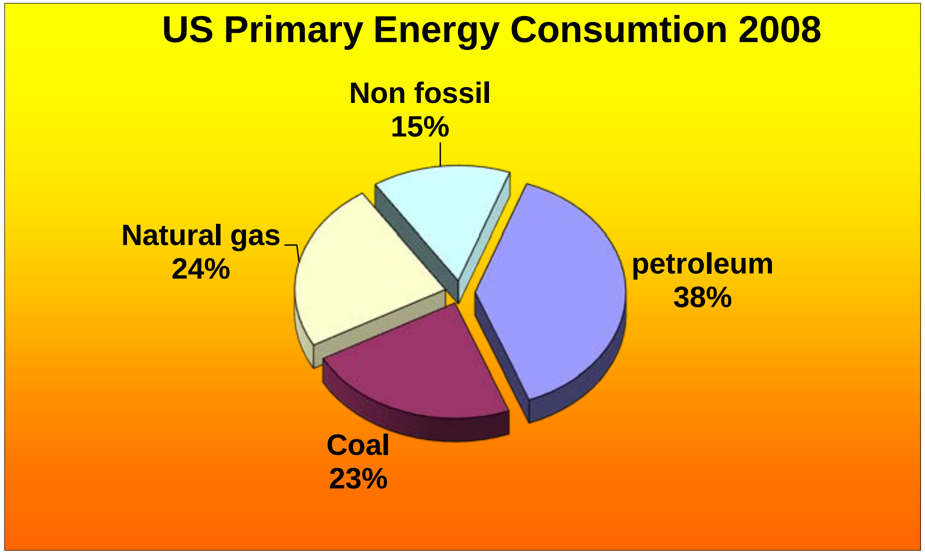
Outline

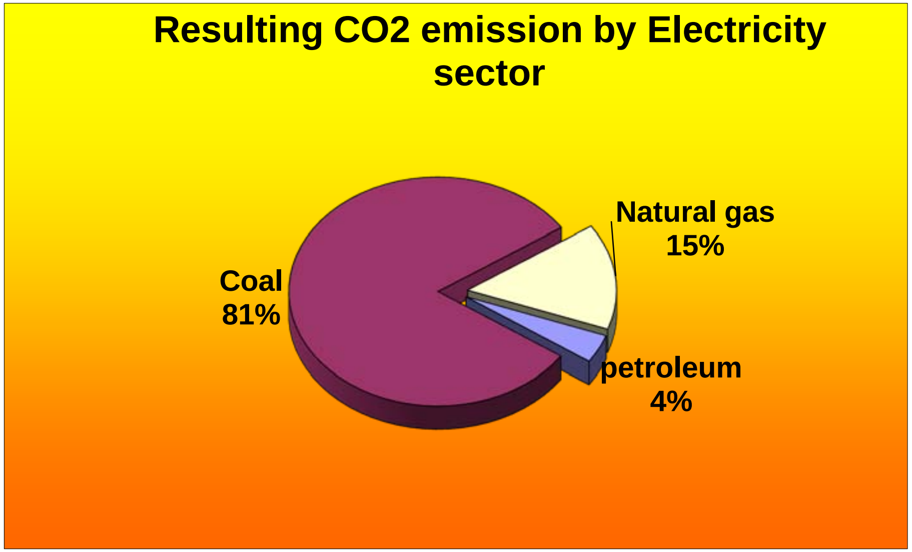
- Motivation and Background
- Objective, Scope and Timeline
- Completed Work and Results
- Future Work
- Questions?

The Drive for Carbon Capture

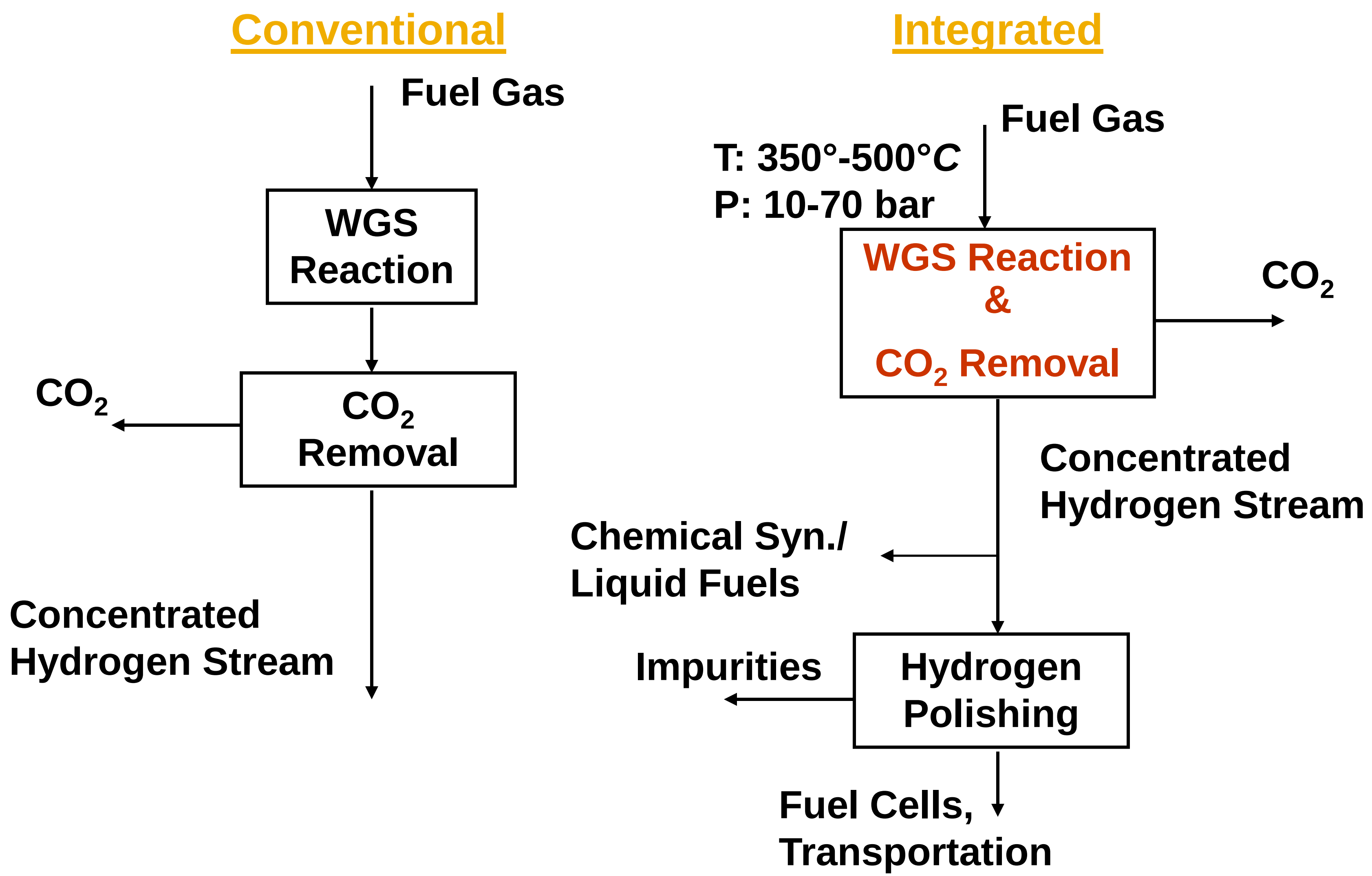








CO_2 Removal and Hydrogen Production



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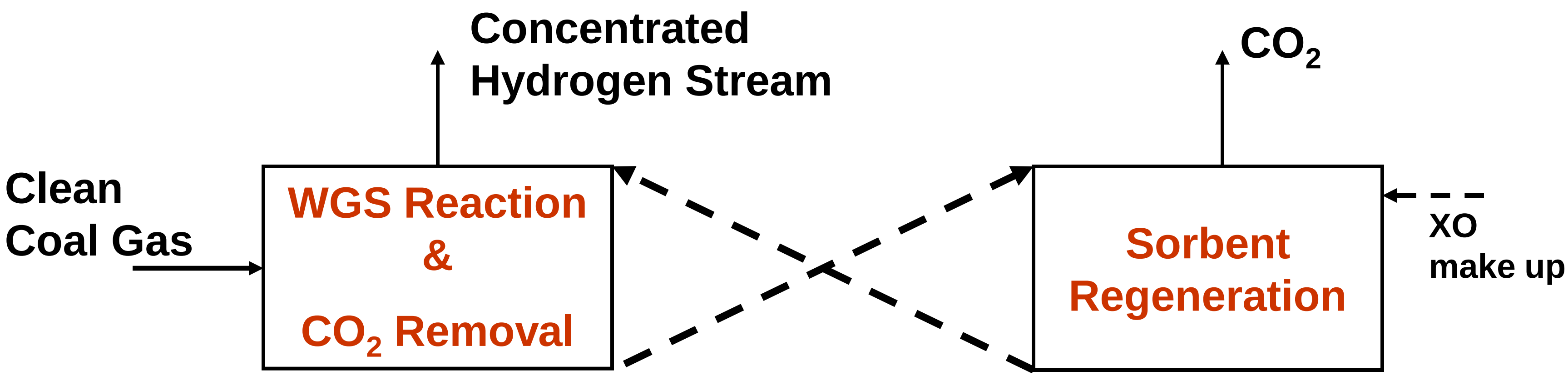
Regenerable Sorbent Approach

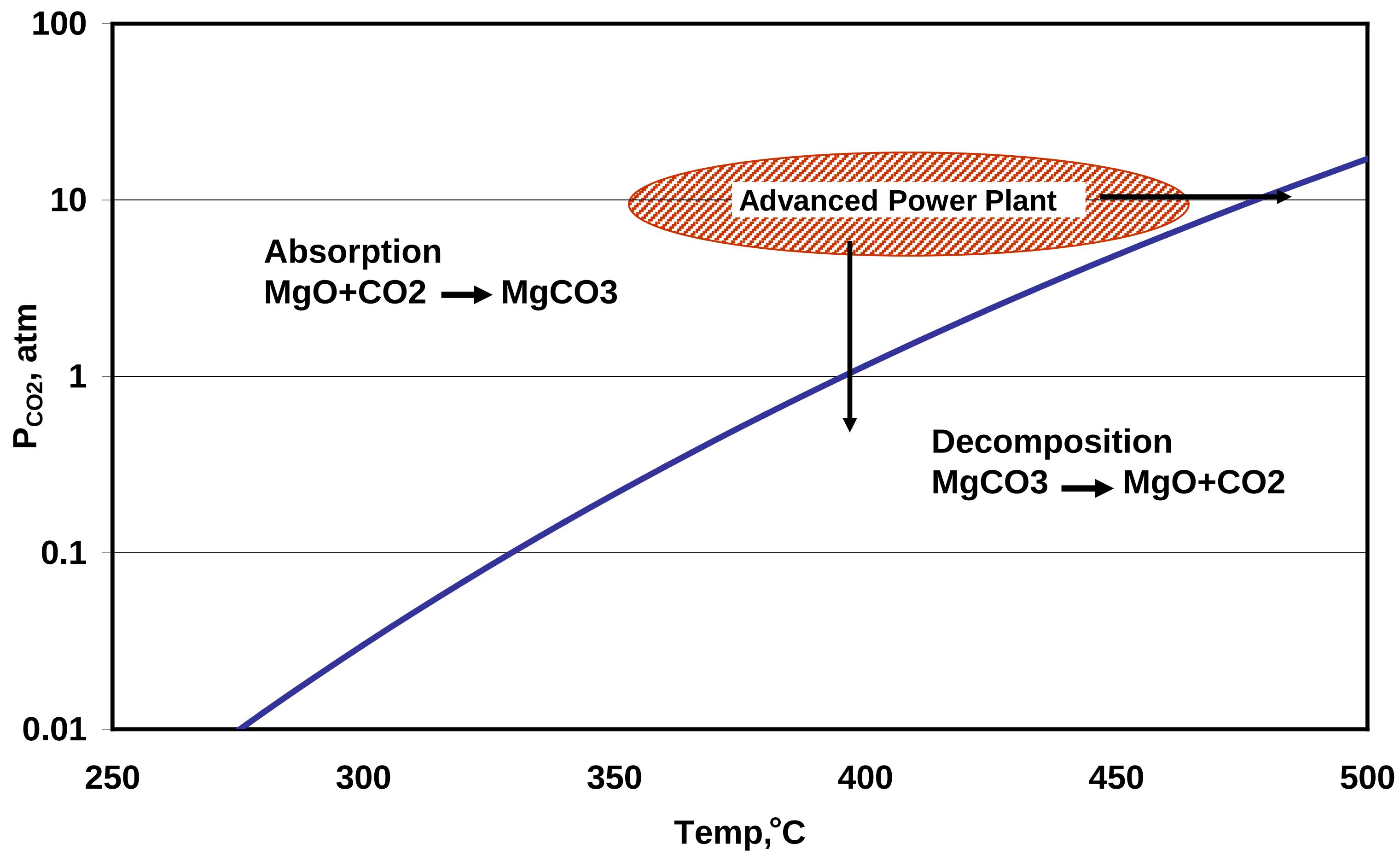


$CO + H_2O \longleftrightarrow CO_2 + H_2$

$XO + CO_2 \longrightarrow XCO_3$

$XCO_3 \longrightarrow XO + CO_2$

MgO-CO_2 Equilibrium



Objective

- The overall objective of this project is to develop a CFD model and to perform Computational Fluid Dynamic (CFD) simulations using Population Balance Equations (PBE) to describe the heterogeneous gas-solid absorption/regeneration and water-gas-shift (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:

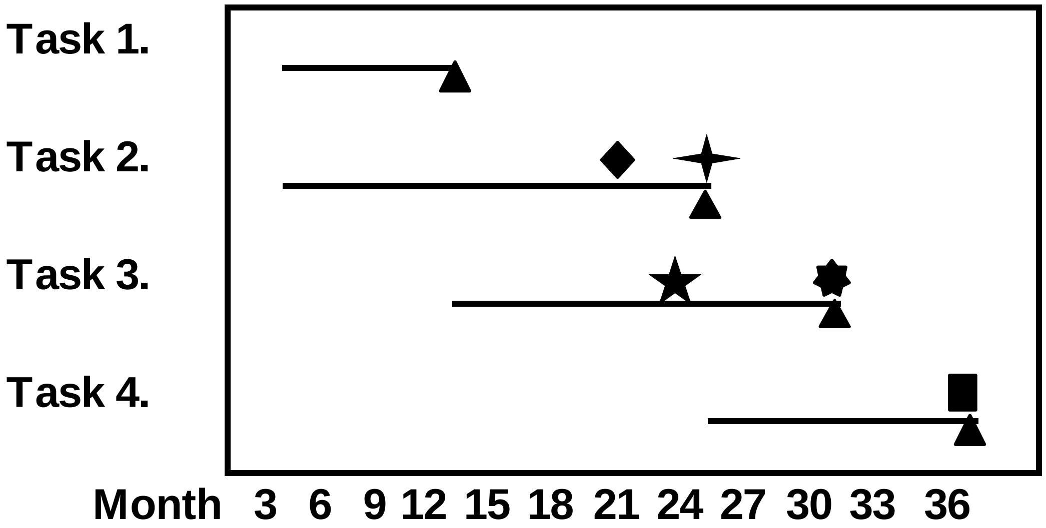
Task1. Development of a CFD/PBE model accounting for the particle (sorbent) porosity distribution and of a numerical technique to solve the CFD/PBE model.

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

Project Schedule

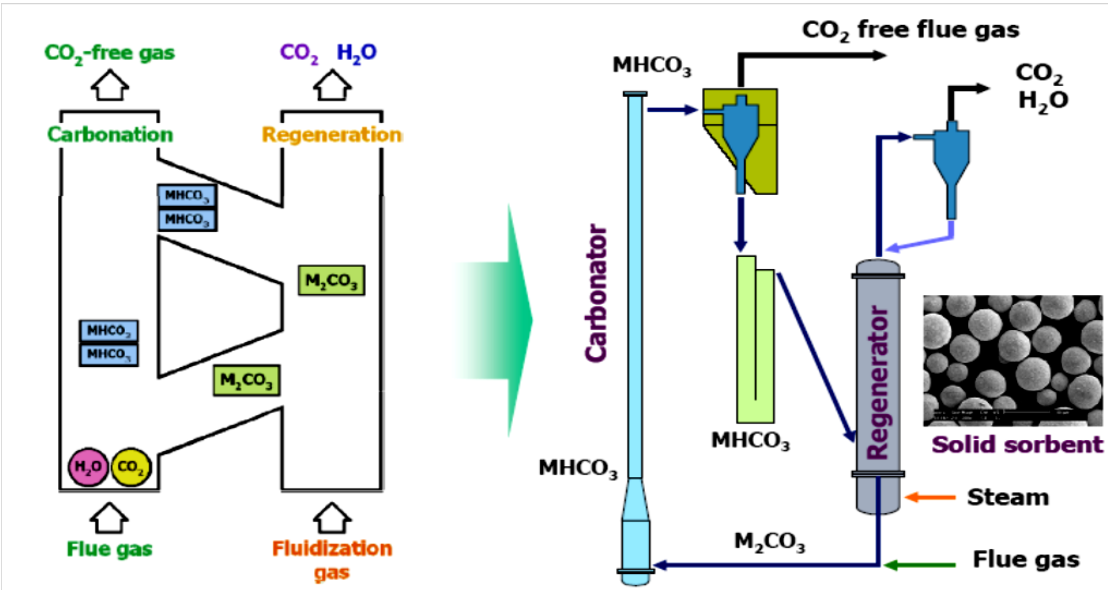


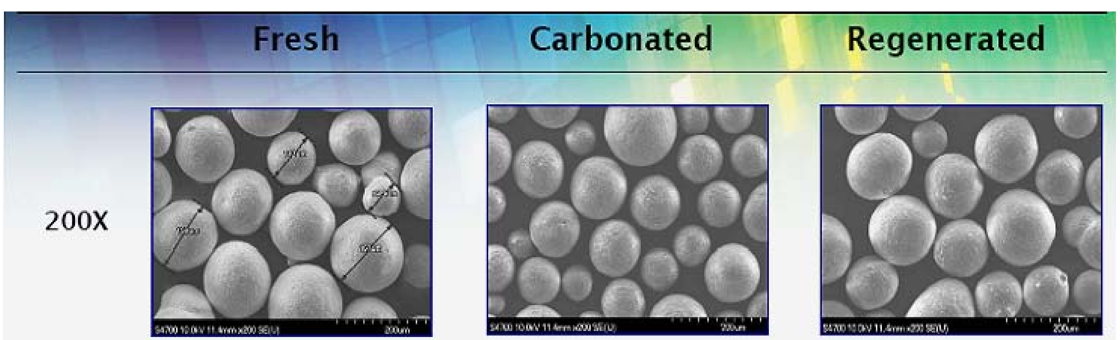
Milestones:

- ▲ 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

Task 1

Development and validation of CFD model



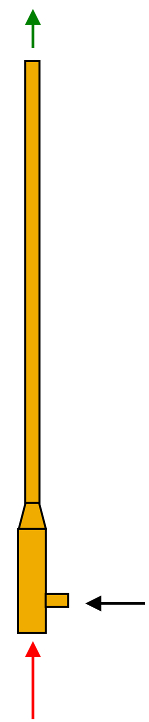


Carbonation:

$$CO_2 + H_2O + K_2CO_3 \rightarrow 2KHCO_3 + \text{Heat}$$

Regeneration:

$$2KHCO_3 \rightarrow CO_2 + H_2O + K_2CO_3 - \text{Heat}$$



Yi et al., International journal of greenhouse gas control, 2007.

Task 1

Numerical Modeling: Conservation Equations

2D, 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

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

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Task 1

Numerical Modeling: Conservation Equations

- 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

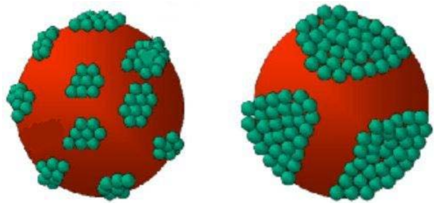
$$\frac{3}{2}[\frac{\partial}{\partial t}(\varepsilon_s \rho_s \theta) + \nabla.(\varepsilon_s \rho_s \theta) v_s] = (-\nabla p_s I + \tau_s) : \nabla v_s + \nabla.(\kappa_s \nabla \theta) - \gamma_s$$

Generation of energy due to solid stress tensor — Diffusion — dissipation

- Reaction Kinetic: Deactivation Kinetic Model (*Park et al, 2007*)

$$-\frac{da}{dt} = k_d C_{CO_2} a$$

$$a = \exp[\frac{[1-\exp(\tau.k_s(1-\exp(-k_d t)))]}{1-\exp(-k_d t)}\exp(-k_d t)]$$



Task 1

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

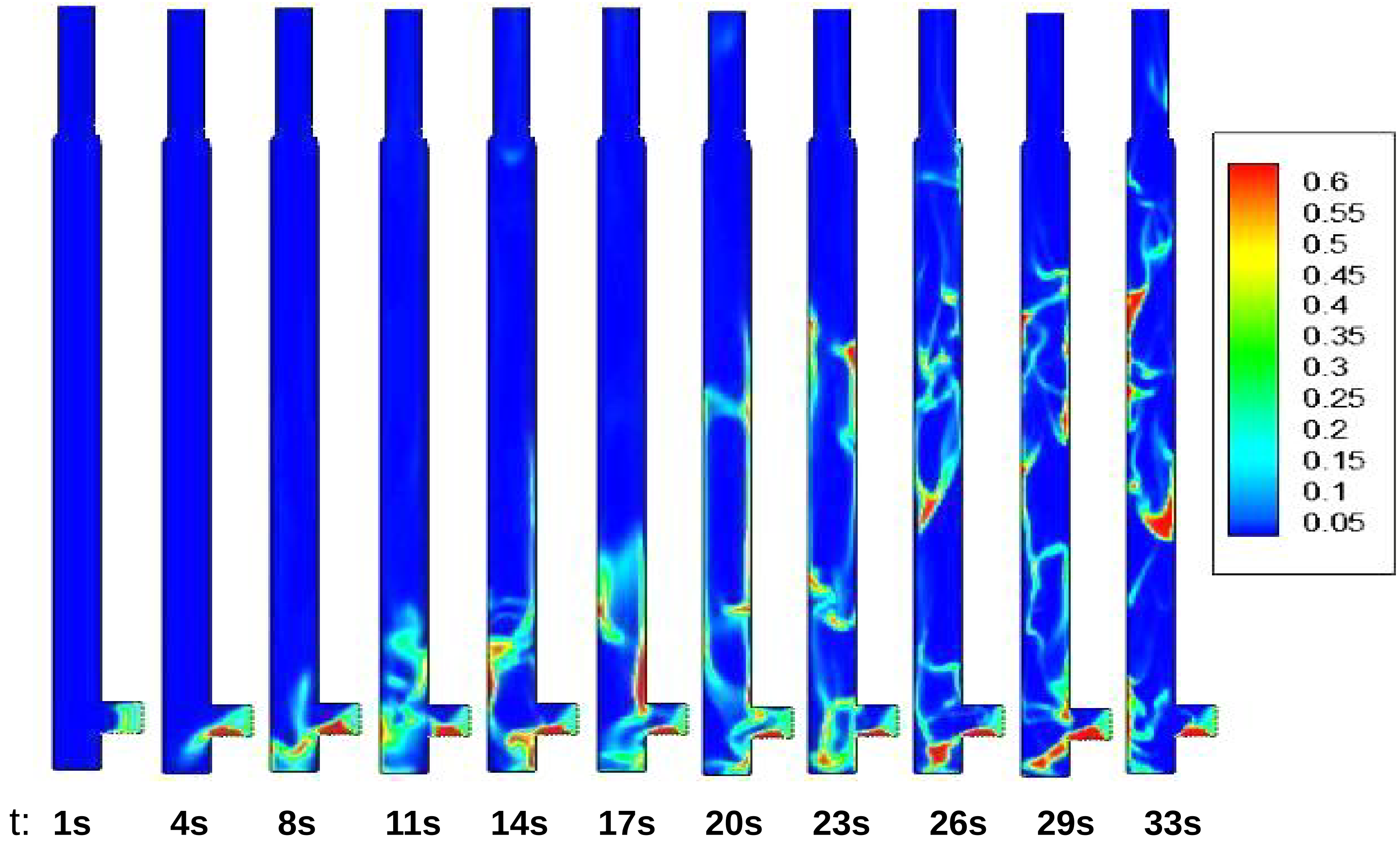
ω < 1

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

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

Task 1

Solid Volume Fraction inside the riser



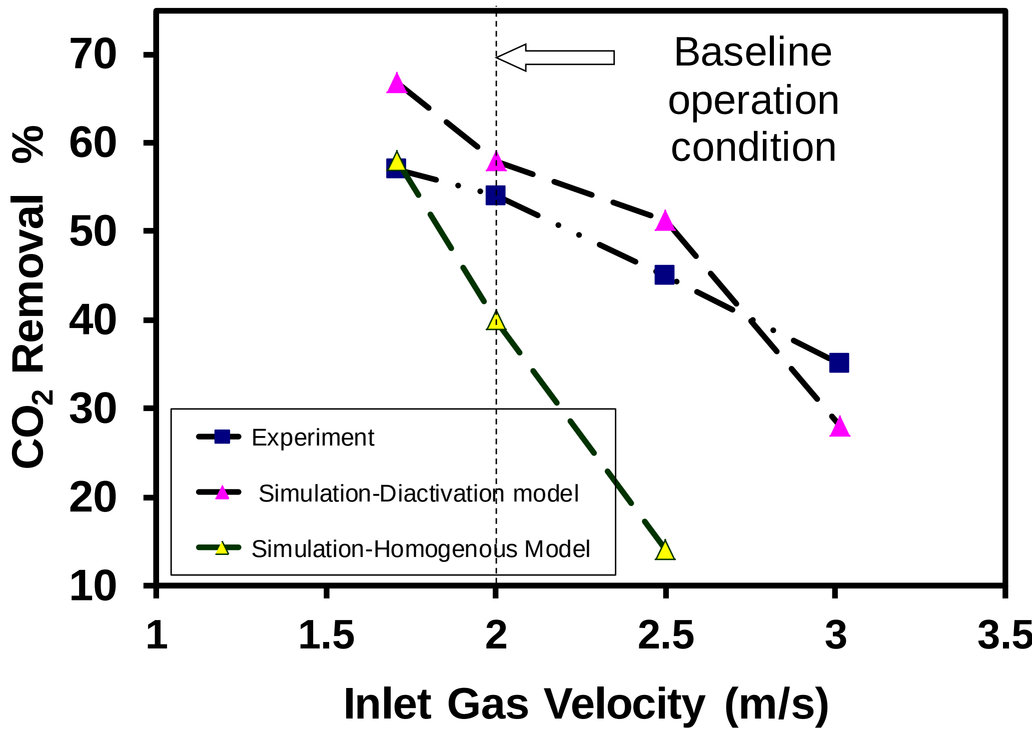
Task 1

Results

Pressure Drop

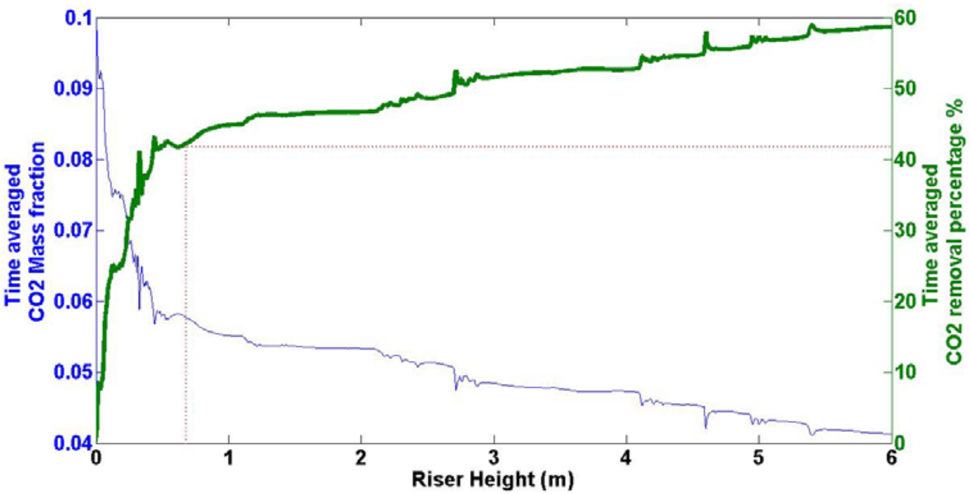
	Time averaged Pressure drop (mm H2O) KIER Experiments	Time averaged Pressure drop (mm H2O) Simulation
DP1	100	107
DP2	200-500	335
DP3	250	270
DP4	70	73

sensitivity to the Inlet Gas Velocity

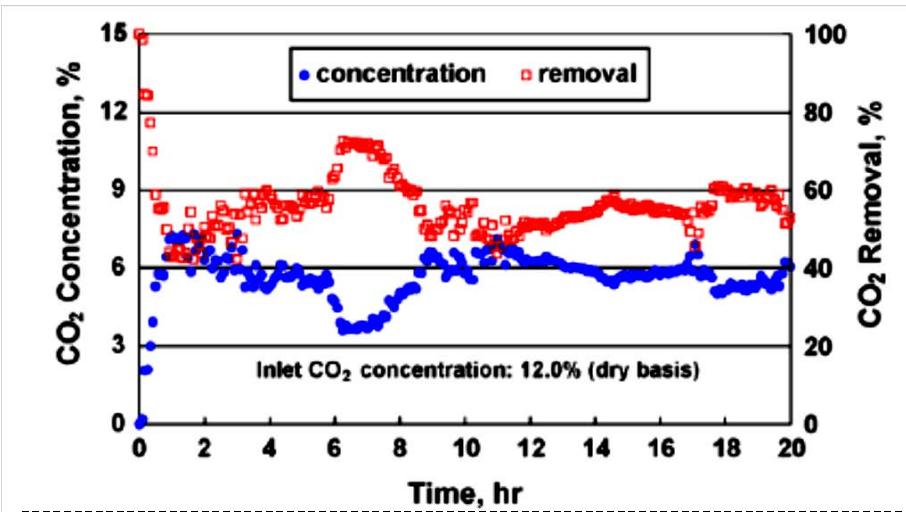


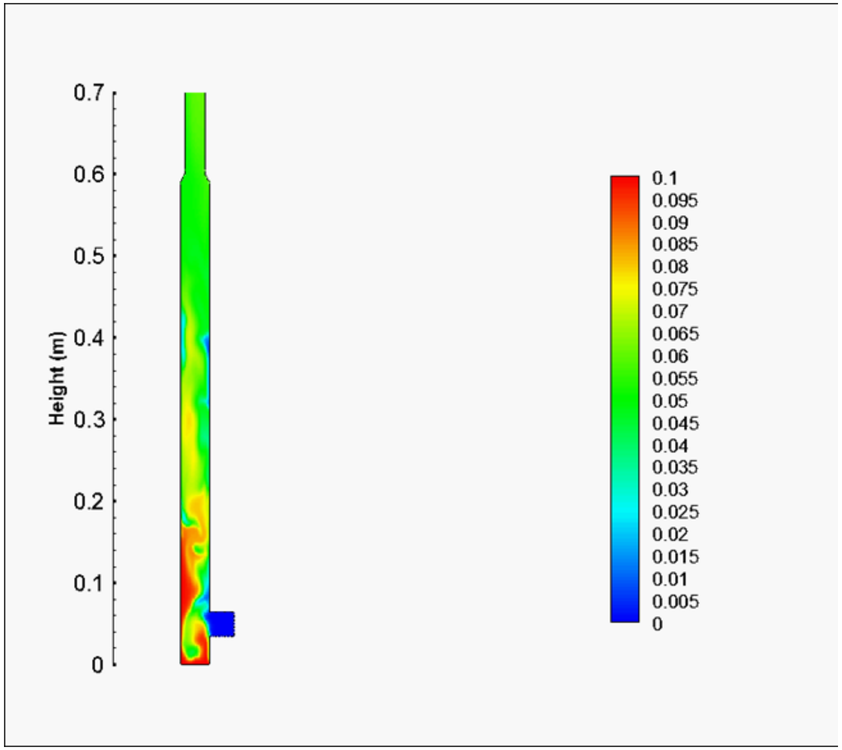
Task 1

Results



$$\%CO_2\ removal = \frac{X_{Co2,in} - X_{Co2}}{X_{Co2,in}} \times 100$$





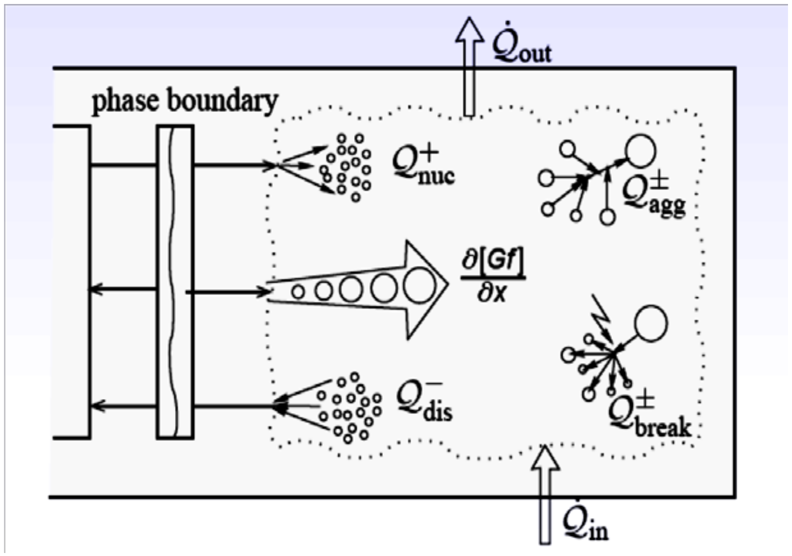
Contours of CO_2 mass fraction

Yi et al., *International journal of greenhouse gas control*, 2007.

Task 1

Formulation of a Population Balance Model (PBM)

What is the Population Balance Equation?



- The population balance equation is a balance equation based on the number density function $f\,(\xi;\,x,\,t)$
- Accounts for the particles accumulating, leaving, entering or being generated or destroyed in a single control volume

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

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

Task 1

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(ξ,x,t)* will be approximated by expansion based on a complete set of trial functions

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

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

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

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Task 1

Moments Transport Equation

$$\boxed{\frac{\partial \mu_i}{\partial t} + \nabla.(\mu_i . v_p) - \nabla.(D'_{pt} \nabla \mu_i) = -(MB + MB_{Conv} + MB_{Diff1} + MB_{Diff2} + MB_{Diff3} + IG)}$$

MB *: Terms due to coordinate transformation (**Moving Boundary**)*

IG*: Contribution due to the **Integration** of **Growth** Term*

Boundary conditions:

$$\frac{d\overline{\xi}_{\min}}{dt} = S_{\min} \quad \text{and} \quad \frac{d\overline{\xi}_{\max}}{dt} = S_{\max}$$

For the application of interest:

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

Task 1

Assumptions

- Uniform and constant particle size distribution.
- Density of the particles is changing during the process due to the reaction between the solid and the gas phase.
- Density distribution function is defined in the range of [ξ_{min}, ξ_{max}] and then using a coordinate transform is changed to [-1, +1].
- Incompressible particle phase .
- Constant maximum sorbent density, corresponding to the completely reacted sorbent.
- Variable minimum sorbent density, corresponding to the fresh sorbent. The rate of change is related to the rate of reaction.
- no breakage or agglomeration in density domain.

Task 1

Implementation and validation of FCMOM method in a CFD code

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- Implementation in Ansys Fluent via UDS

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

$$\phi_s^i = \frac{\mu_i}{\varepsilon_s}$$

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

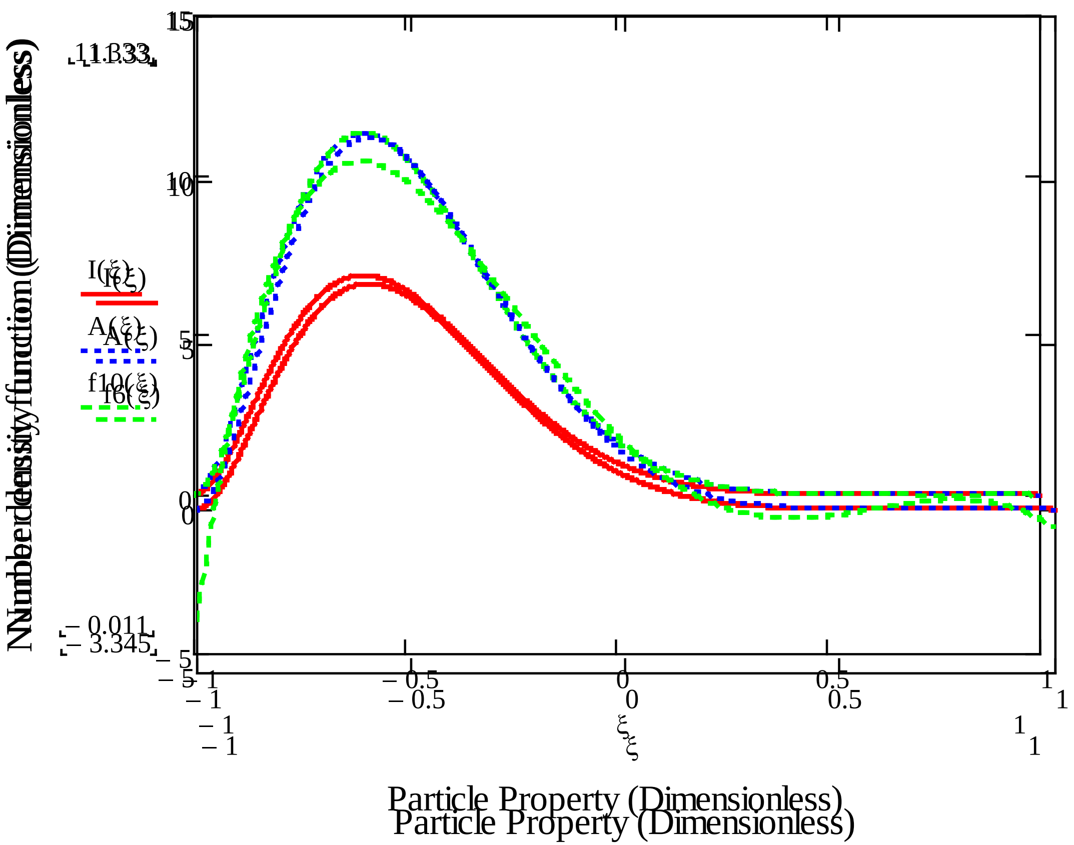
Task 1

Validation

Test case1: Linear Growth, No convection

$$\frac{\partial \mu_i}{\partial t} = \mu_i . K$$

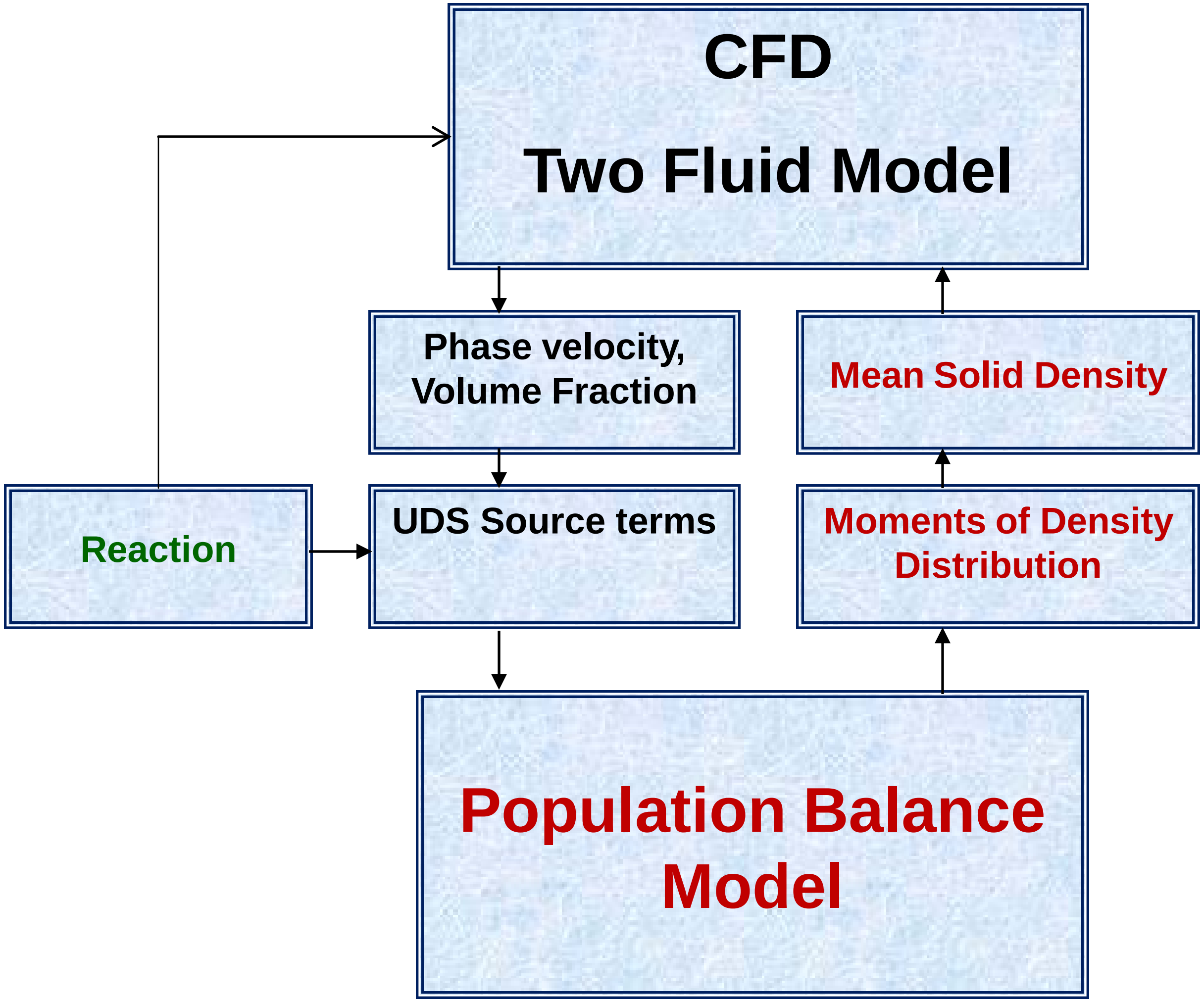
Case study for *k=0.05* and *t=10 sec*



6 moments
10 moments

Task 1

Coupling CFD-PBE



Task 1

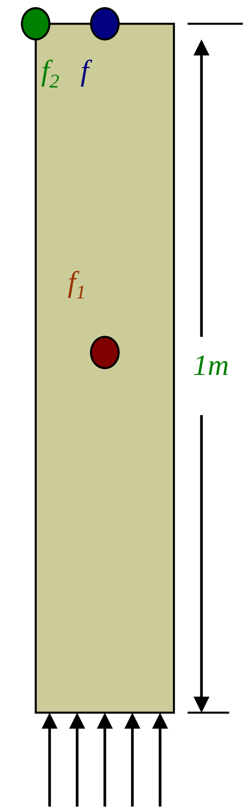
Test case 2: Density Growth (Reaction) and convection

Assumption: Moments are convected with mixture velocity

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

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

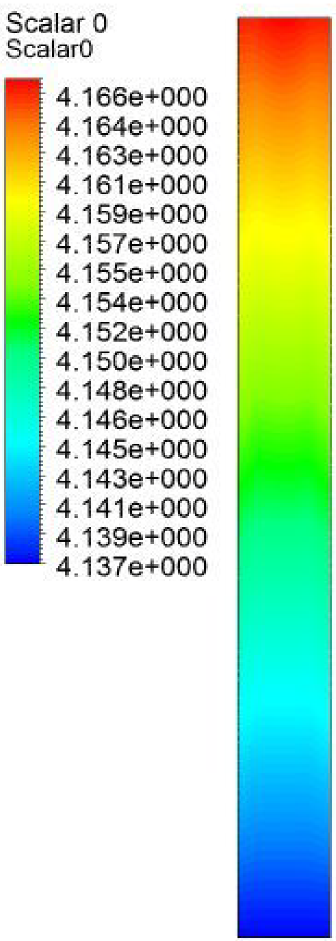
$$
\rho_s = \frac{\left(\frac{\mu_1}{\mu_0}\right)(\xi_{\max} - \xi_{\min}) + (\xi_{\min} + \xi_{\max})}{2}
$$



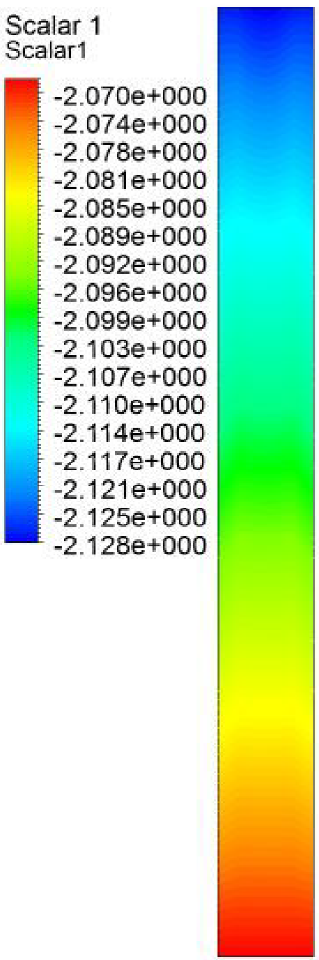
$v_g = v_s = 1$ m/s

$\varepsilon_s = 0.2$

Test case 2: Results

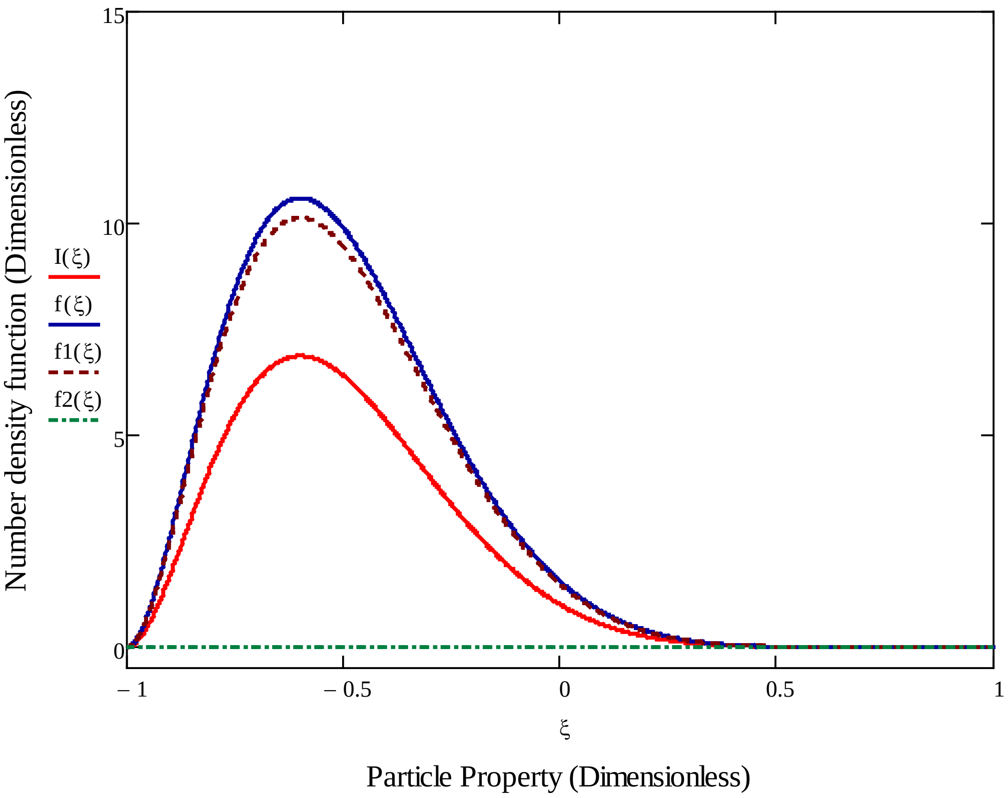


1st Moment



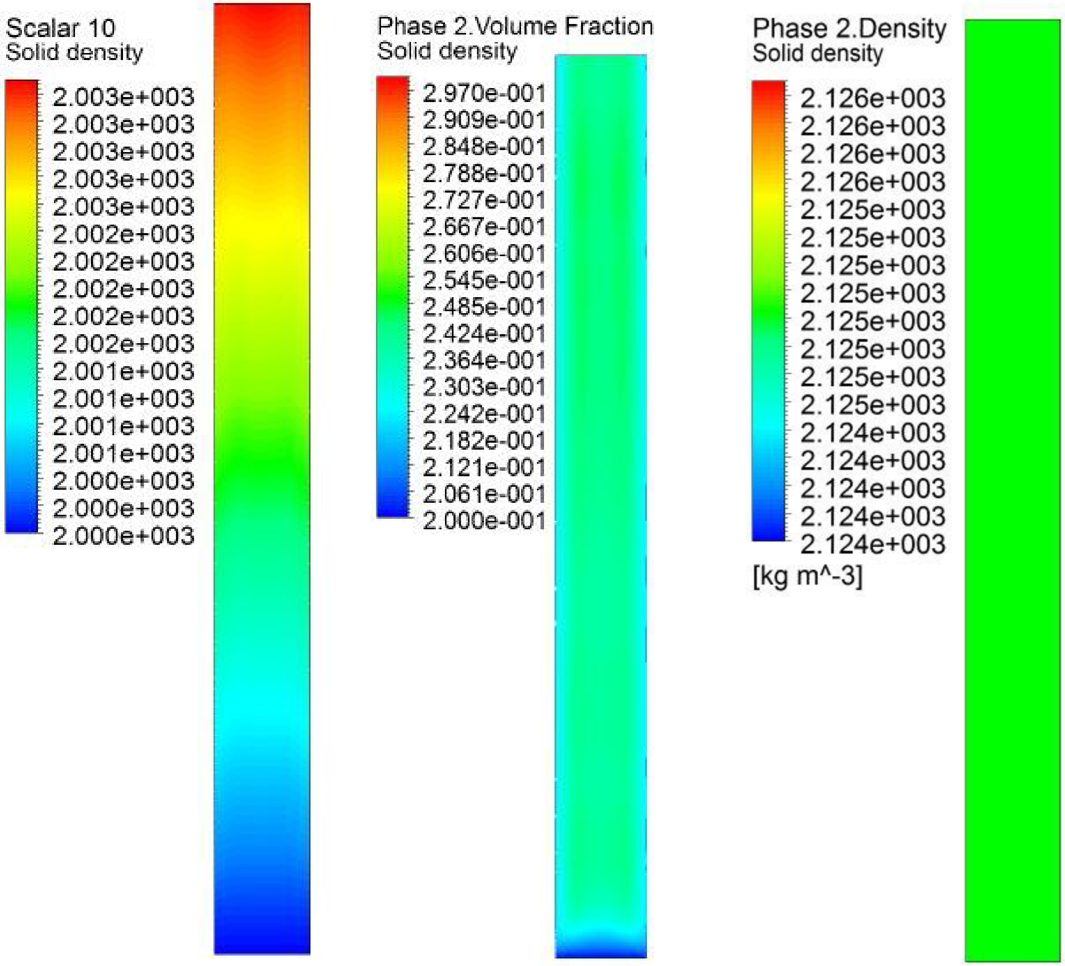
2nd Moment

t = 10s



Test case 2: Results

t = 10s

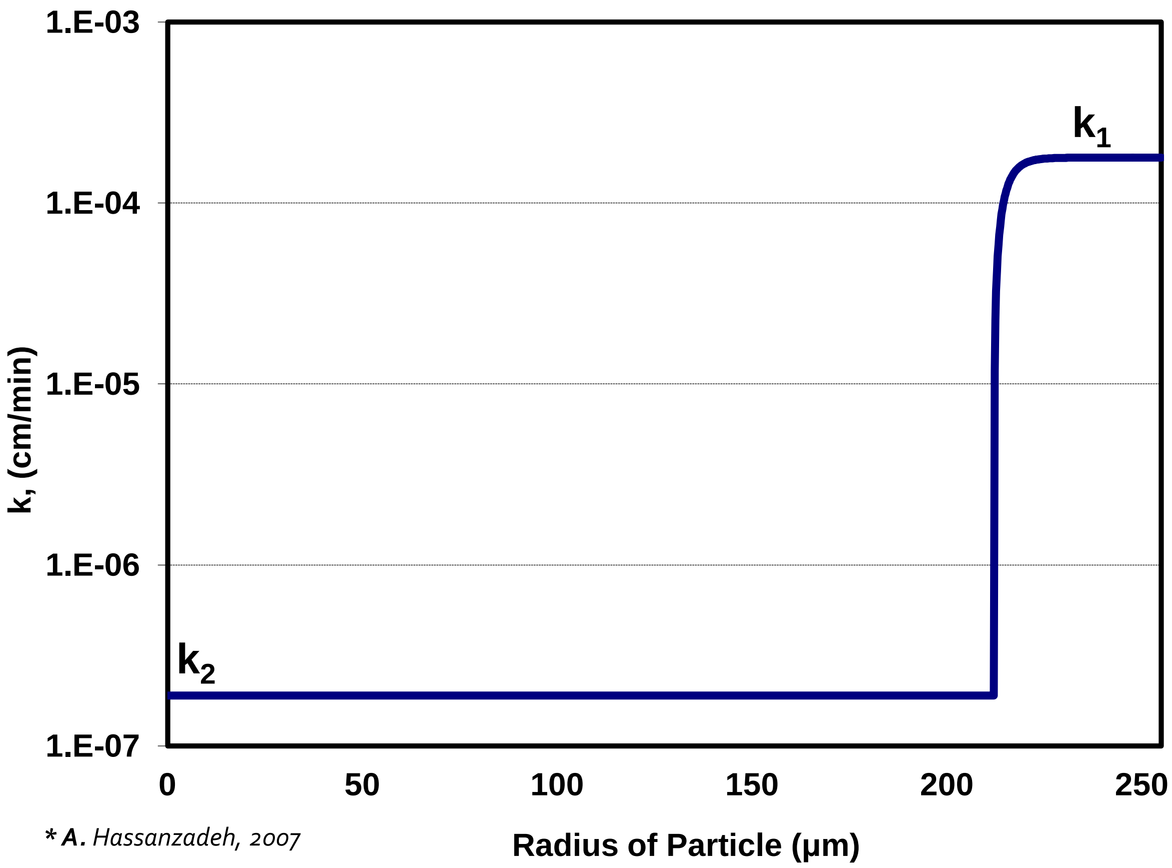


Minimum Density **Solid Volume Fraction** **Solid Density**

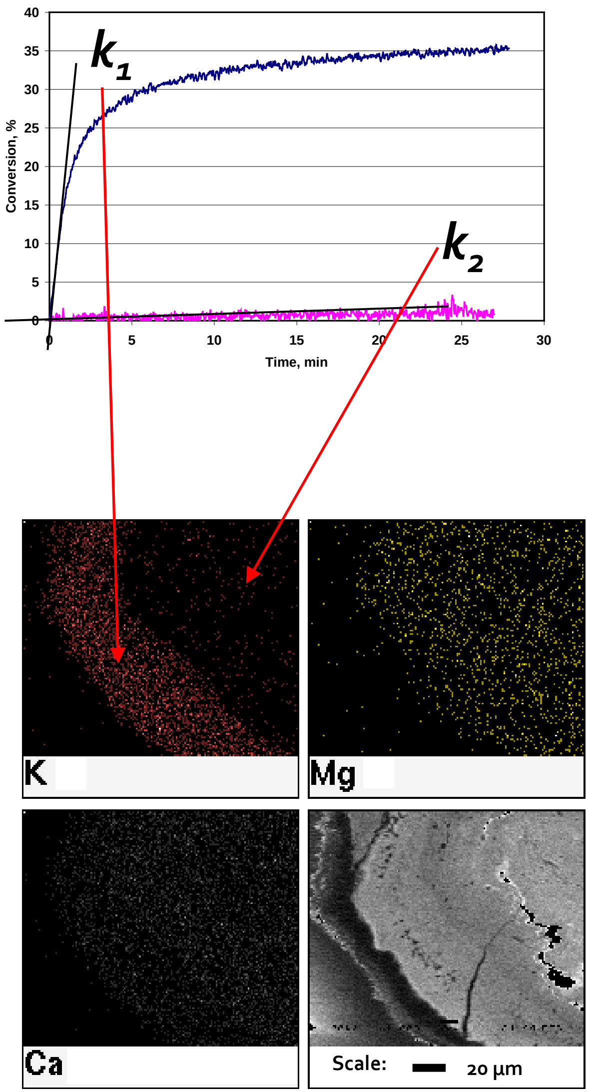
Task 2

Development of a chemical reaction kinetics model

Potassium promoted MgO-based sorbent



* *A. Hassanzadeh, 2007*



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Task 2

Key Assumptions

1- There are two distinct reactive zones inside the particles

2- Process is controlled by both surface reaction and product layer diffusion

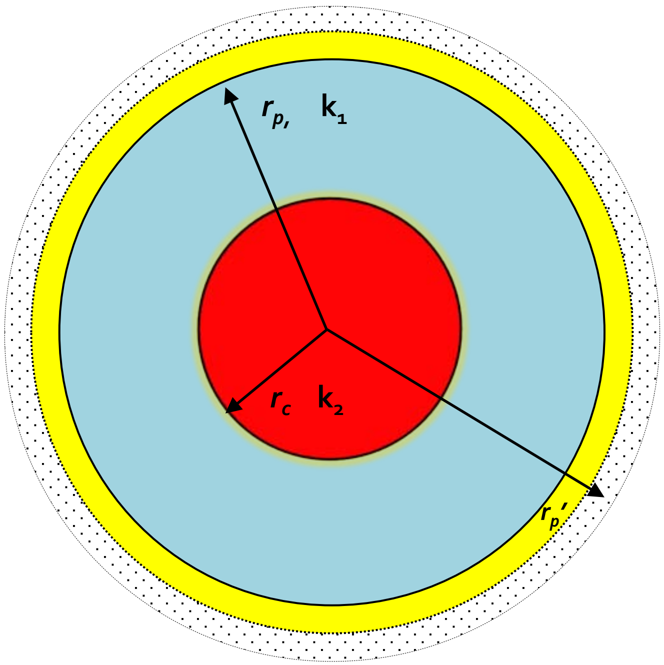
3- There is an Expanding product layer $r_p = r'_p \sqrt[3]{(1 - X) + ZX}$

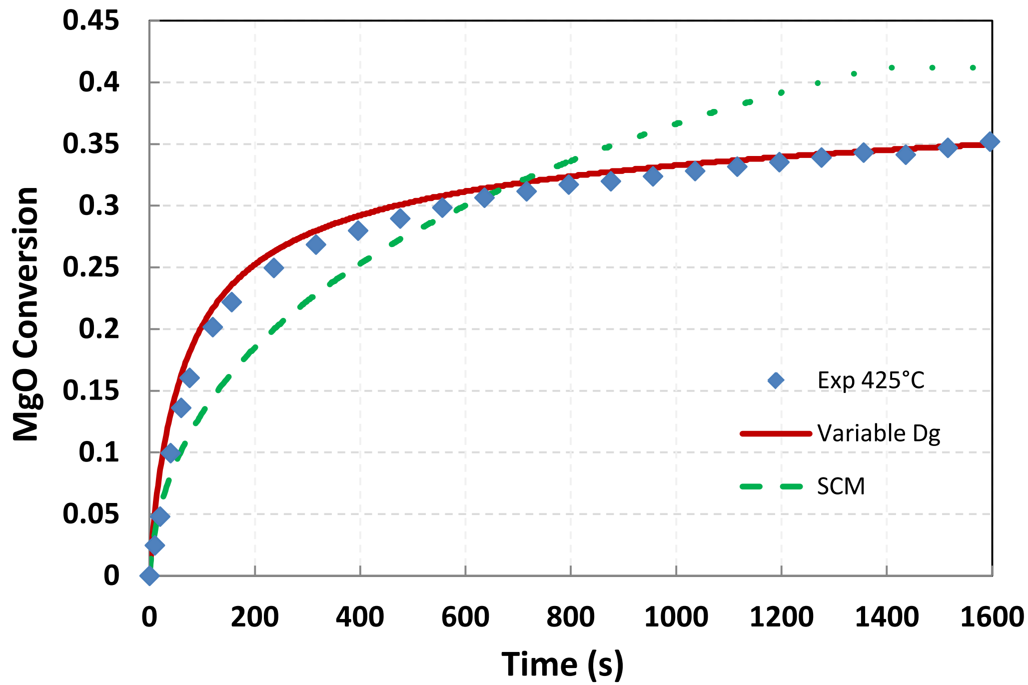
4- D_g is Variable due to the pore closing and is a function of conversion $D_g = D_{g0}(-\alpha X^{\beta})$

5- Intrinsic reaction rate is Arrhenius type $k_s = k_{s0}\exp(-\frac{E}{RT})$

Task 2

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





Gas Film

Product Layer

Highly Reactive Zone (k_1)

Low Reactive Zone (k_2)

r_c : Radius of the low reactive zone (k_2)
r_p : Initial radius of the particles
r_p': Radius of the expanded particle

Task 2

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

$$-\frac{dr_i}{dt} = \frac{(C_b - C_e)/N^o_{MgO}}{\underbrace{\cancel{\frac{r_i^{\,2}}{r_p k_g}}}_{\text{film}} + \underbrace{\frac{(r_p - r_i)r_i}{r_p D_g}}_{\text{Product Layer}} + \underbrace{\frac{1}{k_s}}_{\text{Reaction}}} \Longrightarrow \frac{dr_i}{dt} = -\frac{k_s}{N^o_{MgO}}\left[\frac{(C_b - C_e)}{1 + \frac{k_s}{D_g} r_i (1 - \frac{r_i}{r'_p})}\right]$$

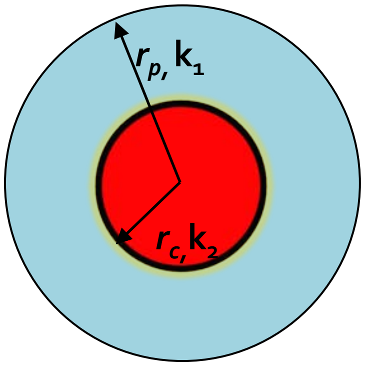
$$D_g = D_{g0}(-\alpha X^{\beta})$$

$$r_p = r'_p \sqrt[3]{(1 - X) + ZX}$$

$$Z = \frac{\rho_{product} \cdot M_{react}}{\rho_{react} \cdot M_{product}}$$

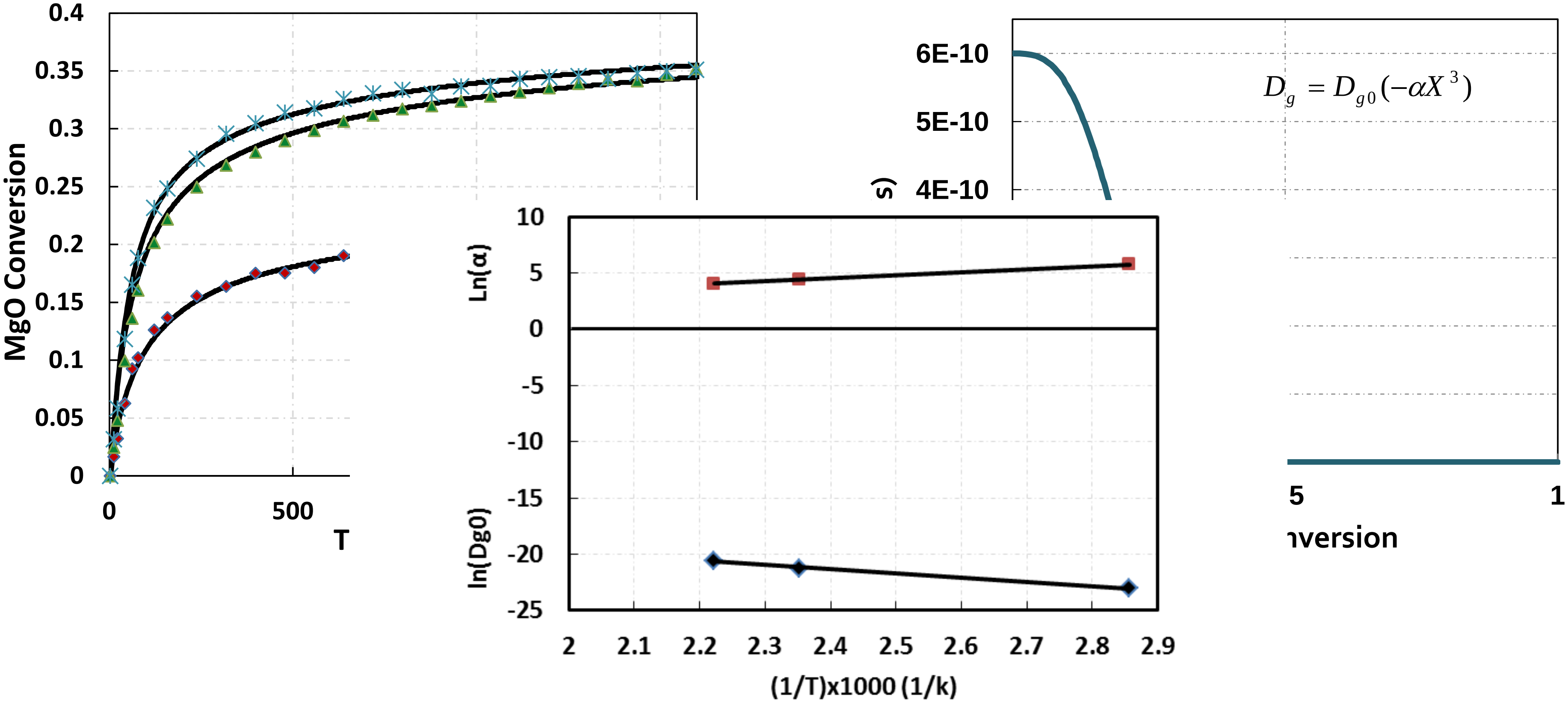
$$\frac{dX}{dt} = -\frac{\frac{3}{r_p}\frac{k_s}{N^o_{MgO}}(C_b - C_e)(1 - X)^{\frac{2}{3}}}{1 + \frac{k_s}{D_g} r_p (1 - X)^{\frac{1}{3}} (1 - \sqrt[3]{\frac{1 - X}{1 - X + XZ}})}$$

$$k_s = \begin{vmatrix} k_1 & for\ \ r \geq r_c \\ k_2 & for\ \ r < r_c \end{vmatrix}$$



Task 2

Reaction Model vs TGA Experimental Date



D_{g0}	1.1E-10	6.0E-10	1.2E-9	2.1E-9
α	3.5E+2	8.1E+1	5.7E+1	4.6E+1
β	3	3	3	3

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Task 2

Validity of Shrinking Core Model

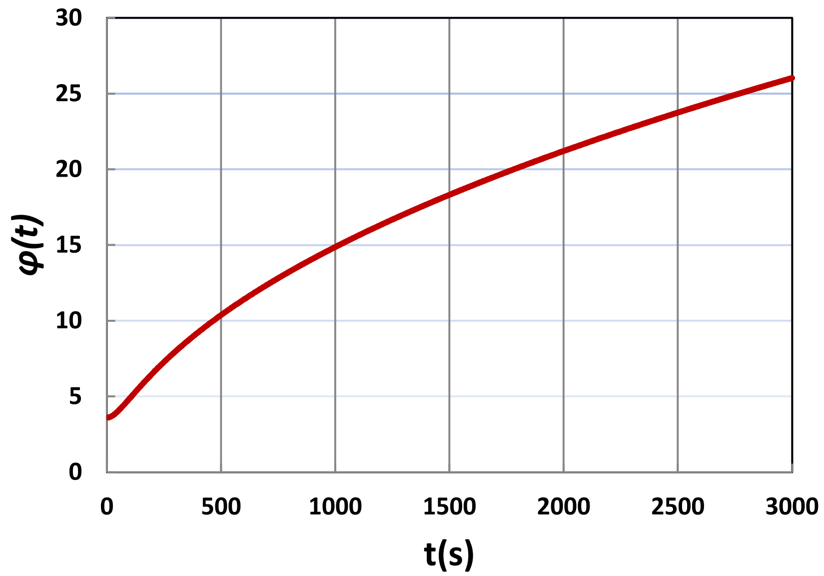
Thiele Modulus

$$\Phi = \sqrt{\frac{Ka^2}{D}} \qquad \frac{reaction}{diffusion}$$

$$\Phi \simeq 0.01 \qquad Reaction\ is\ controlling$$
$$\Phi \simeq 100 \qquad Diffusion\ is\ controlling$$

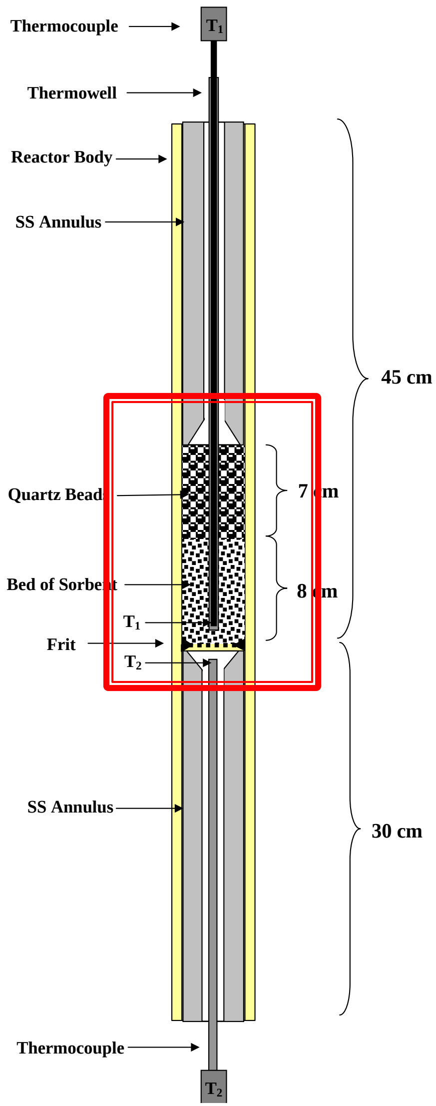
Shrinking core model is applicable in an intermediate regime

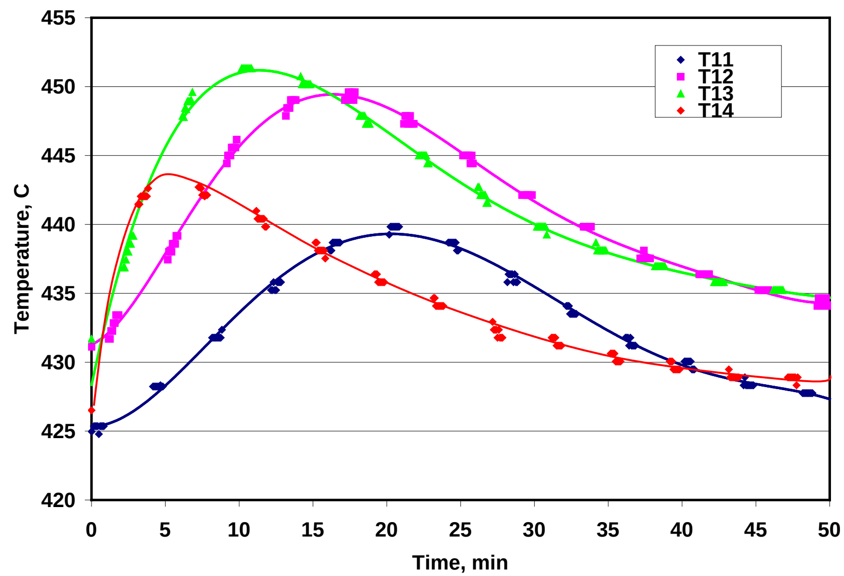
Thiele Modulus in our study

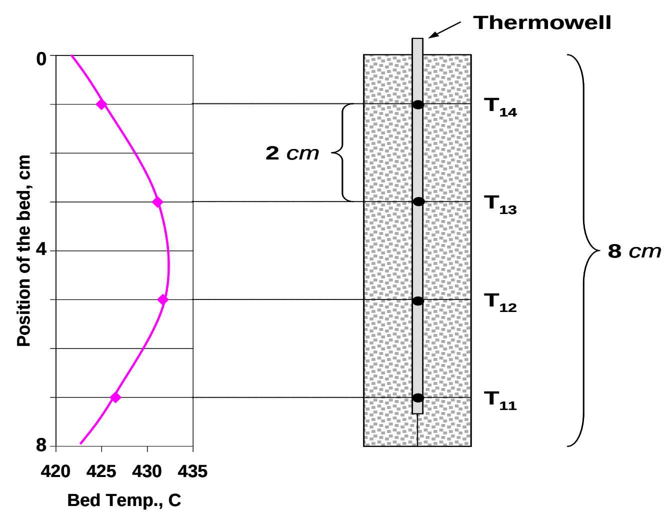


Onischak and Gidaspow, "Separation of Gaseous Mixtures by Regenerative sorption on Porous Solids. Part II: Regenerative separation of CO_2", Recent Developments in Separation Science, ed. N. Li, 1972

Packed-Bed Model

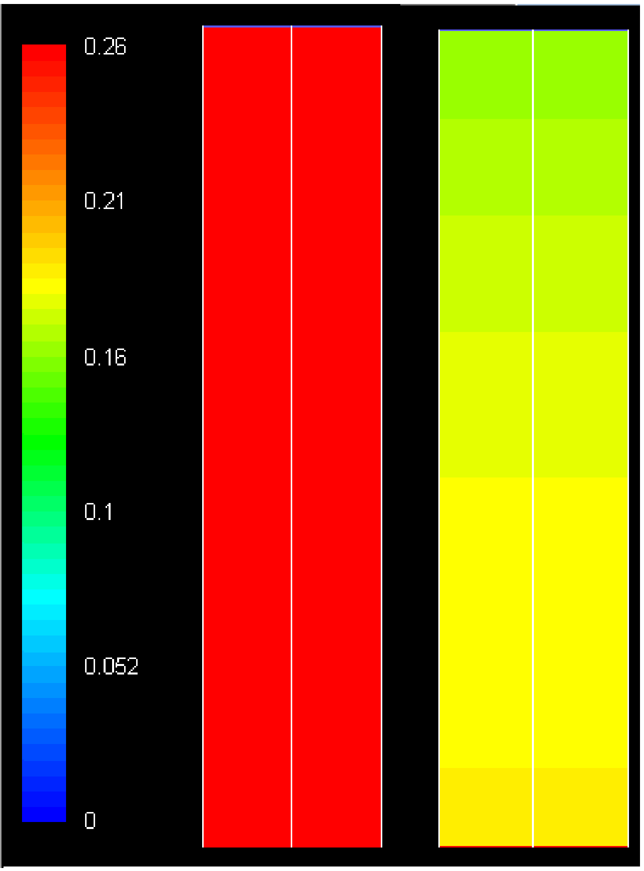


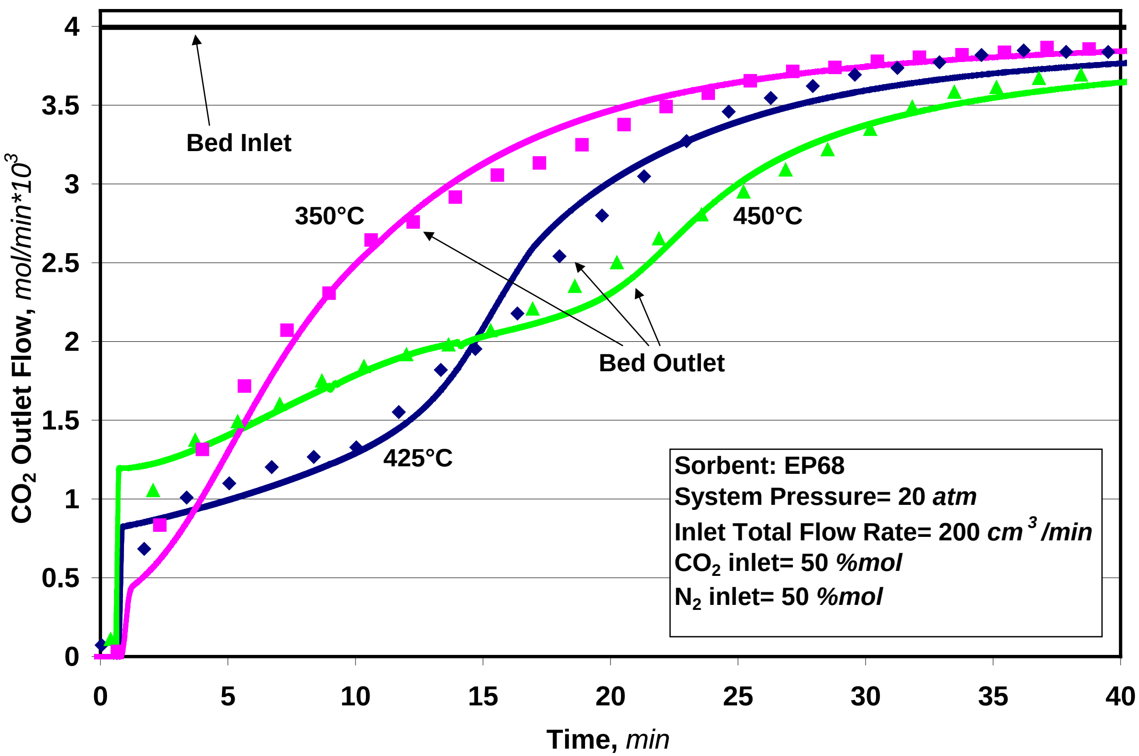




Packed-Bed modeling results

@ 425°C start 50 min





CO_2 Absorption Breakthrough Curve at Different Operating Temperatures

Conclusion

- Results of the CFD model in terms of pressure drops, capturing the cluster formation and CO_2 removal rate is in a good agreement with the experimental data
- An explicit Reaction kinetics model has been developed which is able to explain TGA experimental data very well and is suitable for CFD applications
- PBM and the coupling algorithm for implementation in the CFD code has been developed and verified. More verification is in progress.

Future Work

- Simulations
  - Validation/Verification of the coupled CFD-PBM.
  - Validation of reaction model vs Packed-bed experiments
  - Application of the CFD-PBM in simulation of the circulating fluidized bed reactor
- Experiments
  - Sorbent improvement
  - Reaction rate measurement in shallow/disperse bed reactor

Acknowledgement

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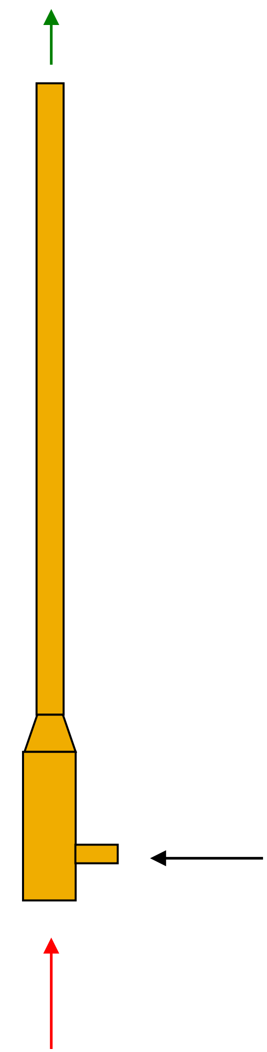
Thanks for your attention

Questions?

Solid inlet	Gas inlet	Outlet	Wall
Solid mass flux = 21 kg/m^2s	Gas velocity= 2 m/s		No slip condition for gas phase
Solid volume fraction = 0.6	Solid volume fraction= 0	P = 1 atm	Partial slip condition for solid phase
Carrier gas mass flux = 0.05 kg/m^2s			
Mass fraction K_2CO_3 = 0.35 Mass fraction $KHCO_3$ = 0 Mass fraction Inert = 0.65	Mass fraction CO_2 = 0.1 Mass fraction H_2O = 0.15 Mass fraction N_2 = 0.75		



A second order discretization scheme was used to discretize the governing equation
domain including 34x1200 uniform rectangular cells.