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CFD SIMULATION OF CO₂ SORPTION IN A CIRCULATING FLUIDIZED BED USING THE DEACTIVATION KINETIC MODEL

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ABSTRACT

The Computational Fluid Dynamics (CFD) approach was used to simulate sorption of CO₂ using solid sorbents in the riser section of a circulating fluidized bed. The simulation results were compared with the experimental data of Korea Institute for Energy Research (KIER) for continuous CO₂ sorption using potassium carbonate in a circulating fluidized bed system.

INTRODUCTION

Coal-based power plants generate more than 50 percent of the today's United States electric power (1). This means coal will continue to play a significant role in electricity generation for the foreseeable future. Therefore, the global emission of CO₂ and its impact on climate change will continue to increase. Separation and sequestration of CO₂ has been investigated by many researchers during the past decade including pre-combustion CO₂ separation (gasification), oxyfuel combustion, and post-combustion CO₂ separation. Post-combustion CO₂ separation, which includes chemical and physical sorption of CO₂ from flue gases, is a challenging process due to the low pressure and low concentration of CO₂ in flue gas which requires high volumetric flow rates of flue gas to be processed. In addition, these processes generally use sorbents to capture the CO₂ and these sorbents need to be regenerated and used continuously in the process. The regeneration of sorbents is an energy demanding process that reduces the overall efficiency of the power plant. Therefore, developing more efficient and economically feasible processes for CO₂ removal has been one of our goals in recent years. Based on National Energy Technology Laboratory's 2009 report (1), the goal for energy consumption of novel sorbents should be one third of energy consumption for today's commercially available sorbents, which are basically amine-based liquid sorbents.

Recent studies have shown that alkali-metal-based solids could be a promising sorbent for efficient and cost-effective CO₂ removal from combustion gases (2, 3, 4). Ryu and coworkers (3) studied sodium- and potassium-based sorbents and concluded that they possess excellent features like superior attrition resistance, high CO₂ sorption capacity, and high bulk density. Afterward, their group (5,6) at Korea Institute for Energy Research (KIER) used a potassium-based solid sorbent to

perform two sets of experiments in a 2 Nm³/hr and a 100 Nm³/hr facility for CO₂ capture from flue gas. They found that using a circulating fluidized bed (CFB) ensures the continuous CO₂ removal process from dilute flue gases at laboratory and bench scales.

To scale up this CFB process, a state-of-the-art design tool based on CFD simulation is needed. However, to this point, few detailed simulations of this process have been conducted (7, 8) that can capture qualitatively the behavior of the system.

In this study, we used experimental data provided by KIER (5) to validate our CFD simulation for the CO₂ capture process in the riser part of a circulating fluidized bed using a potassium-based solid sorbent.

EXPERIMENT USED FOR SIMULATION

For our simulation we used the experiments of Yi *et al.* (5), which includes a circulating fluidized bed consisting of a riser as carbonator and a bubbling fluidized bed as regenerator. Figure 1 shows the schematic of CO₂ sorption using a circulating fluidized bed system.

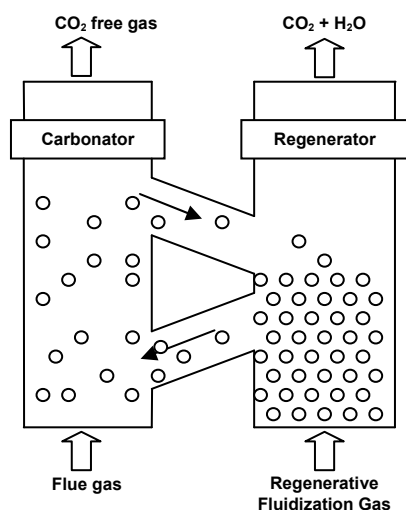
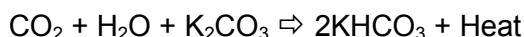
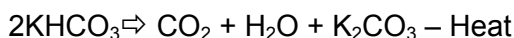


Figure 1. Schematic of CO₂ sorption process in a CFB

Flue gas containing CO₂, H₂O, and N₂ enters the carbonator and reacts with fresh (regenerated) solid sorbent containing 35% K₂CO₃ as shown below:



CO₂-free gas exits from the top of the riser while reacted sorbents go back to the regenerator to react with steam according to the following reaction:



A very slender 2.5 cm ID and 6 m height riser, with an expanded 3.5 cm ID mixing zone at the first 0.6 m of the bottom of the riser as the carbonator and a 10 cm ID and 1.28 m height bubbling bed as the regenerator reactor, were used. In this study, our focus has been on the simulation of the riser and the carbonation process.

The operating condition is atmospheric pressure and 80° C in the riser. The flue gas inlet velocity was 2 m/s with 12% CO₂ (dry basis) and 12.3% H₂O composition. The solid circulating rate was controlled using a solid valve and was set to 21 kg/m².s as the baseline operating condition.

The potassium-based Sorb KX35 sorbent has a bulk density of 1100 kg/m³ and particle density of 2394 kg/m³ with average particle size of 98 μm. The attrition index (AI) of this sorbent has been reported as 0.1% at 10 std l/min (3).

Differential pressure was measured at four different elevations along the riser. In addition, CO₂ concentration at the riser outlet was monitored continuously.

NUMERICAL MODELING

The CFD simulation of this work is based on a two-dimensional Eulerian-Eulerian approach in combination with the kinetic theory of granular flow (9, 10). To convert the real geometry to a reduced two-dimensional domain, the solid mass flux was kept constant as the basis for calculation.

The assumptions of our numerical simulation include the isothermal condition for the process, and consider the gas phase as an ideal gas and the particles in the solid phase to be of uniform and constant size and density. Fluent 6.3 code was used to solve a set of governing equations including:

Mass conservation

$$\text{For gas phase: } \frac{\partial}{\partial t}(\varepsilon_g \rho_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{v}_g) = \dot{m}_g$$

$$\text{For solid phase: } \frac{\partial}{\partial t}(\varepsilon_s \rho_s) + \nabla \cdot (\varepsilon_s \rho_s \mathbf{v}_s) = \dot{m}_s$$

Momentum conservation

For gas phase:

$$\frac{\partial}{\partial t}(\varepsilon_g \rho_g \mathbf{v}_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{v}_g \mathbf{v}_g) = -\varepsilon_g \nabla P + \nabla \cdot \boldsymbol{\tau}_g + \varepsilon_g \rho_g \mathbf{g} - \beta_{gs}(\mathbf{v}_g - \mathbf{v}_s)$$

For solid phase:

$$\frac{\partial}{\partial t}(\varepsilon_s \rho_s \mathbf{v}_s) + \nabla \cdot (\varepsilon_s \rho_s \mathbf{v}_s \mathbf{v}_s) = -\varepsilon_s \nabla P + \nabla \cdot \boldsymbol{\tau}_s + \varepsilon_s \rho_s \mathbf{g} + \beta_{gs}(\mathbf{v}_g - \mathbf{v}_s)$$

Species conservation

$$\text{For gas phase: } \frac{\partial}{\partial t}(\varepsilon_g \rho_g y_i) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{v}_g y_i) = R_j \quad i=1, 2, 3$$

$$\text{For solid phase: } \frac{\partial}{\partial t}(\varepsilon_s \rho_s y_i) + \nabla \cdot (\varepsilon_s \rho_s \mathbf{v}_s y_i) = R_j \quad i=1, 2, 3$$

And conservation of solid phase fluctuating energy:

$$\frac{3}{2} \left[\frac{\partial}{\partial t}(\varepsilon_s \rho_s \theta) + \nabla \cdot (\varepsilon_s \rho_s \theta \mathbf{v}_s) \right] = (-\nabla p_s I + \boldsymbol{\tau}_s) : \nabla \mathbf{v}_s + \nabla \cdot (\kappa_s \nabla \theta) - \gamma_s$$

Where κ_s and γ_s are conductivity of fluctuating energy and collisional dissipation of solid fluctuating energy, respectively.

For the gas-solid inter-phase exchange coefficient β_{gs} there are different correlations available in the literature. Garg et al. (7) showed that using an EMMS-based model as proposed by Li et al. (11) gives better results compared to the drag model proposed by Gidaspow (12). Nikolopoulos et al. (13) showed that the EMMS model increased the accuracy of the simulation at the lower part of the riser, resulted in better prediction for solid concentration and pressure distribution, and was able to account for heterogeneous solid structures and cluster formation in the riser.

The EMMS-based drag model has been used in this study as follows:

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

Where, $\omega(\varepsilon_g)$ is called the heterogeneity factor and is defined as

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

And

$$C_{D0} = \frac{24}{Re_p} (1 + 0.15 Re_p^{0.687}) \quad \text{for } Re_p < 1000$$

$$C_{D0} = 0.44 \quad \text{for } Re_p > 1000$$

In addition, $k-\varepsilon$ turbulent model has been used to take care of turbulent fluctuations of the gas-solid mixture.

Initially, there was no solid in the riser and the concentration of CO₂ was zero as well. The summary of the boundary conditions is shown in Table 1.

A second order discretization scheme was used to discretize the governing equation throughout the domain including 34x1200 uniform rectangular cells. In order to check the grid independence of the solution, the computations were also performed using two other coarser grids. The comparison between the calculated pressure drop using 34x1200 and 17x600 uniform grids showed an insignificant difference in the calculated pressure drop along the riser.

REACTION KINETIC MODEL

There is very little information available on the kinetics of the carbonation reaction of K₂CO₃ in the literatures. Onischak and Gidaspow (14) have proposed a first order homogenous reaction kinetic model which is dependent only on CO₂ concentration and is independent of sorbent and H₂O concentration. Recently, Park et al. (15) investigated different kinetic models including a Homogenous Model (HM), Shrinking Core Model (SCM), and Deactivation Model (DM). They concluded that the Deactivation Model explains fixed bed reactor experimental data better than the other two above-mentioned models.

Garg et al. (7) used the proposed HM to simulate a similar process. Although their simulation was able to capture the CO₂ concentration at the outlet of the riser, the simulation results were approximately three times more sensitive to the changes in

gas flow rate than observed in their experiments. It seems that the independence of the reaction model to gas velocity is one of the issues of HM.

The Deactivation Model (DM) proposed by Park et al. includes gas volumetric flow rate in the kinetic model. The concept of this model is based on analogy between deactivation of catalyst particles by coke formation and deactivation of sorbent particles by carbonation.

Table 1. Summary of Boundary Conditions for Baseline Operating Condition

Solid inlet	Gas inlet	Outlet	Wall
Solid mass flux = 21 kg/m ² s	Gas velocity= 2 m/s	P = 1 atm	No slip condition for gas phase
Solid volume fraction = 0.6	Solid volume fraction= 0		Partial slip condition for solid phase
Carrier gas mass flux = 0.05 kg/m ² s			
Mass fraction K ₂ CO ₃ = 0.35 Mass fraction KHCO ₃ = 0 Mass fraction Inert = 0.65	Mass fraction CO ₂ = 0.1 Mass fraction H ₂ O = 0.15 Mass fraction N ₂ = 0.75		

IN the deactivation Model, the effect of the formation of a product layer on the surface of sorbent particles (which results in an additional diffusion resistance and reduction in available active surface area) is lumped into a reducing activation factor with an exponentially deactivation rate.

$$R = k C_{CO_2} a$$

Where a is activity of the sorbent and defined as:

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

τ , called surface time, is defined as the ratio of available pore surface to the volumetric flow rate of flue gas. $k_s = 2.44 \times 10^{-3} \text{ m/s}$ and $k_d = 1.42 \times 10^{-4} / \text{s}$ are surface reaction constant and deactivation constant, respectively. (15)

RESULTS

The simulation results for CO₂ removal percentage and sensitivity analysis of the model to the changes in gas flow rate and pressure drop along the riser are presented in this section. Simulations ran for 200 seconds of the processing time and, as it takes about 90 seconds for the solid inventory in the riser to become stable, the first 100 seconds of the simulations were not considered in the calculation of the time averaged results. Our results are being compared with the experimental data of KIER provided by Yi et al. (5).

Figure 2 shows the simulated axial profile of time averaged CO₂ mass fraction and CO₂ removal percentage at the different elevations in the riser at the baseline operating condition.

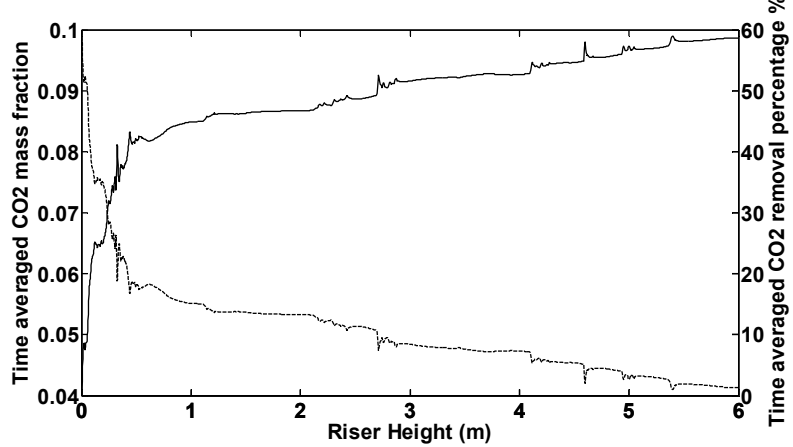


Figure 2. Simulated time averaged CO₂ mass fraction and CO₂ removal percentage

The CO₂ removal percentage at the outlet of the riser is 58%, which is very close to the reported 54% removal in the experimental data.

In addition, the axial profile of the time averaged CO₂ mass fraction showed that around 60% of CO₂ removal takes place in the first 0.6 m of rise (mixing zone),

which is due to the higher solid concentration and solid circulation in this region.

Furthermore, the effect of gas flow rate on the percentage of CO₂ removal has been investigated. The results showed that the increasing inlet gas flow rate (inlet gas velocity) decreases the CO₂ removal percentage, which is in-line with the experimental data. The higher gas flow rate means shorter residence time, which results in reduction of surface time, τ , in DM and, in turn, increases the deactivation factor and decreases the CO₂ removal (see Figure 3). Moreover, our model was able to capture the sensitivity of the CO₂ removal process to the gas flow rate much better than Garg et al. (7) who used the homogeneous reaction model. Sensitivity of CO₂ removal to the changes in the solid circulation rate was also investigated. As was expected, increasing the solid circulation rate resulted in increased CO₂ removal

Table 2. Differential Pressure at Different Elevations

	Differential Pressure (mm H ₂ O) KIER Experiments Yi et al (5)	Differential Pressure (mm H ₂ O) Simulation
DP1	100	107
DP2	200-500	335
DP3	100-210	270
DP4	70	67

percentage. Similar to the experimental data and the simulation of Garg et al. (7), the results of our simulation were also sensitive to the variations in the solid circulation rate. However, due to some inconsistencies in the reported data by Yi et al. (5), no comparison with their data was made in this study.

Table 2 shows the comparison between the pressure drop predicted using our simulation with the experimental results of Yi et al. (5) at 4 different elevations of 0.52 m, 2.27 m, 4.07 m, and 5.87 m which are referred to as DP1, DP2, DP3, and DP4, respectively. Our simulation closely predicted the pressure drop over the DP1, DP2, and DP4 sections

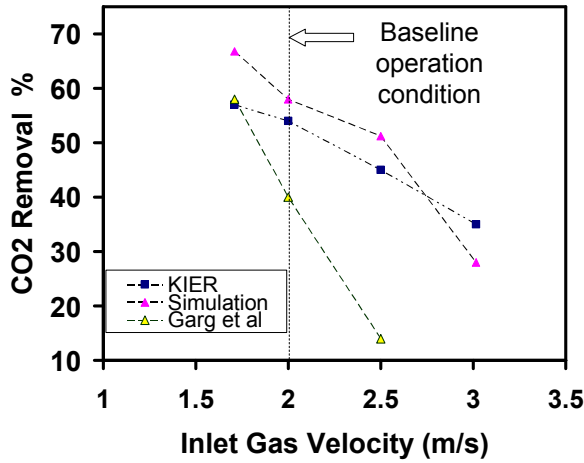


Figure 3. Effect of inlet gas velocity on CO₂ removal percentage

and over predicts the pressure drop over the DP3 section. This could be due to a couple of reasons. First, in the 20 hours of pressure drop profiles reported by KIER, there is a sudden reduction over the DP2 and DP3 sections at around the 11th hour of operation that remains up to the end of the experiment. This reduction in pressure drop is apparently due to an undisclosed change in the operating condition

that cannot be implemented in our simulation. The second possible reason could be due to the inaccuracy of the EMMS drag model in predicting a wide range of solid phase concentration and heterogeneities.

CONCLUSION

A 2-D Eulerian-Eulerian CFD simulation based on kinetic theory of granular flow in combination with deactivation kinetic model has been used to simulate the KIER experiments for CO₂ sorption in a circulating fluidized bed using solid sorbent. The simulation was able to predict CO₂ removal percentage at the riser outlet at different gas flow rate in good agreement with experimental data. Our simulation was also able to predict the pressure drop in the riser and dependency of the CO₂ conversion to gas flow rate in line with the KIER experimental data.

NOTATION

$ a$	Activation coefficient (-)	y_i	Species i mass fraction
$ C_{D0}$	Drag coefficient (-)	γ_s	Collisional dissipation of solid fluctuating energy (kg/s-m ³)
d_p	Particle diameter (m)	κ_s	Conductivity of solid fluctuating energy (kg/m-s)
k_d	Deactivation rate constant (1/s)	θ	Granular temperature (m ² /s ²)
k_s	Surface reaction constant (m/s)	β_{gs}	Inter-phase drag coefficient (kg/s-m ³)
\dot{m}	Inter-phase mass transfer (kg/m ³ -s)	ρ_i	Density of phase i (kg/m ³)
R_j	Heterogeneous reaction rate (kg/m ³ -s)	τ_i	Stress tensor of phase i (Pa)
u_i	Superficial velocity of phase i (m/s)	ε_i	Volume fraction of phase i (-)
v_i	Local velocity of phase i (m/s)	τ	Surface time (s/m)

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