

Effect of a diffuser on gas-solid behavior in CFB riser for CO<sub>2</sub> capture<sup>†</sup>Hoanju Yoo<sup>1</sup>, Hokyuu Moon<sup>1</sup>, Hwimin Seo<sup>2</sup>, Yong Ki Park<sup>2</sup> and Hyung Hee Cho<sup>1,\*</sup><sup>1</sup>Department of Mechanical Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul 120-749, Korea<sup>2</sup>Green Chemistry Process Research Division, Korea Research Institute of Chemical Technology, 141 Gajeongro, Yuseong, Daejeon 305-600, Korea

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## Abstract

Gas-solid fluidized beds have been used in CO<sub>2</sub> capture processes because of their high mixing characteristic and heat and mass transfer. Sufficient residence time of solid particles in a reactor is required to capture CO<sub>2</sub>. However, a fraction of solid particles pass through a reactor without capturing CO<sub>2</sub> due to normal reaction characteristics. Therefore, the objective of the present study was to increase the sorbent residence time using a diffuser in a reactor for CO<sub>2</sub> capture. An Eulerian-Eulerian model in a commercial CFD program was employed to simulate gas-solid flow in the reactor. First, sensitivity analysis depending on operating conditions was conducted to predict the residence time of solid particles. The diffuser was located in the middle of the reactor and the angle of the diffuser was changed. Solid particles dispersed in the radial direction because of gas characteristics in the diffuser and increased the residence time. The results showed that the diffuser increased the sorbent residence time, so that the probabilities of gas-solid reaction would be also improved.

**Keywords:** CO<sub>2</sub> capture; Circulating fluidized bed riser; Diffuser; Residence time

## 1. Introduction

CO<sub>2</sub> capture is the most fundamental technology in carbon capture and sequestration, and is broadly classified into three types: Post-combustion, pre-combustion and oxyfuel-combustion. A post-combustion process can also be categorized as wet-solvent, dry-sorbent, and membrane separation depending on the method of CO<sub>2</sub> treatment [1]. Among them, the dry-sorbent method employs a well-known gas-solid fluidized bed, and has unique advantages, such as that it produces minimal wastewater, sorbent materials are inexpensive, and the method operates over a wide range of temperatures [2]. However, CO<sub>2</sub> capture using a gas-solid fluidized bed still requires a large amount of energy for operation. To commercialize carbon capture and sequestration using a gas-solid fluidized bed, additional energy penalties need to be reduced.

As part of these efforts, Park et al. (2013) proposed a multi-stage CO<sub>2</sub> capture system to reduce the regeneration energy of spent-sorbent [3]. Fig. 1 shows the basic concept of the process [4]. The process is composed of three different operating temperature stages: Low, mid and high. Dry sorbents circulate between the adsorption reactor and the regeneration reactor in each stage. This system uses reaction heat when capturing CO<sub>2</sub> as required thermal energy for regeneration of spent-sorbent, i.e. reaction heat transfers from the adsorption reactor

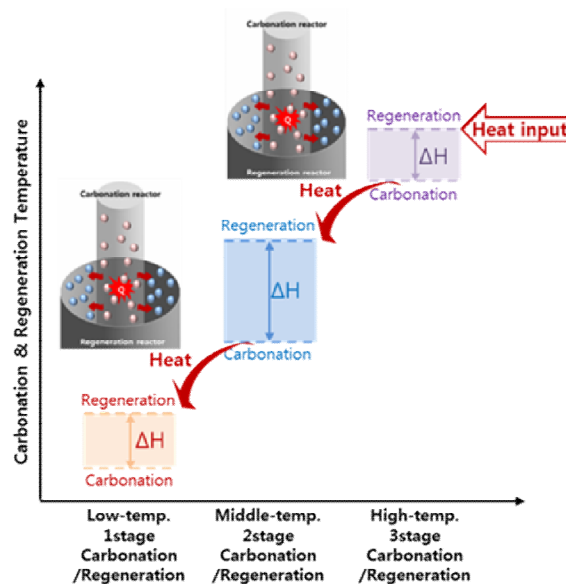


Fig. 1. Schematic of the multi-stage CO<sub>2</sub> capture system [4].

of the high temperature stage to the regeneration reactor of the mid temperature stage. A heat supply is only required for a high temperature regeneration reactor, so that it can handle a large amount of flue gas at a relatively small energy cost.

However, for a gas-solid heterogeneous reaction for CO<sub>2</sub> capture, dry sorbent should stay in the carbonation reactor in about 30 seconds to one minute [5, 6]. A multi-stage CO<sub>2</sub>

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Table 1. A summary of the governing equations and constitutive equations [7, 8].

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Mass conservation for the gas and solid phase

$$\text{Gas: } \frac{\partial}{\partial t}(\alpha_g \rho_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g) = 0 \quad (1)$$

$$\text{Solid: } \frac{\partial}{\partial t}(\alpha_s \rho_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s) = 0 \quad (2)$$


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Momentum conservation for the gas and solid phase

$$\text{Gas: } \frac{\partial}{\partial t}(\alpha_g \rho_g \vec{v}_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g \vec{v}_g) = -\alpha_g \nabla p + \alpha_g \rho_g \vec{g} + \nabla \cdot \alpha_g \vec{\tau}_g - K_{gs}(\vec{v}_g - \vec{v}_s) \quad (3)$$

$$\text{Solid: } \frac{\partial}{\partial t}(\alpha_s \rho_s \vec{v}_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \vec{v}_s) = -\alpha_s \nabla p - \nabla p_s + \alpha_g \rho_g \vec{g} + \nabla \cdot \alpha_s \vec{\tau}_s + K_{gs}(\vec{v}_g - \vec{v}_s) \quad (4)$$


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k - ε dispersed turbulence model

$$\text{Kinetic energy: } \frac{\partial}{\partial t}(\alpha_g \rho_g k_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g k_g) = \nabla \cdot \left( \alpha_g \frac{\mu_{t,g}}{\sigma_k} \nabla k_g \right) + \alpha_g G_{k,q} - \alpha_g \rho_g \epsilon_g - K_{sg} (2k_g - \sqrt{2k_g} \sqrt{3\theta_s}) \quad (5)$$

$$\text{Rate of dissipation: } \frac{\partial}{\partial t}(\alpha_g \rho_g \epsilon_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g \epsilon_g) = \nabla \cdot \left( \alpha_g \frac{\mu_{t,g}}{\sigma_\epsilon} \nabla \epsilon_g \right) + \alpha_g \frac{\epsilon_g}{k_g} (C_{1\epsilon} G_{k,g} - C_{2\epsilon} \rho_g \epsilon_g) - C_{2\epsilon} \frac{\epsilon_g}{k_g} K_{gs} (2k_g - \sqrt{2k_g} \sqrt{3\theta_s}) \quad (6)$$


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Transport equation of the granular temperature

$$\frac{3}{2} \left[ \frac{\partial}{\partial t}(\alpha_s \rho_s \theta_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \theta_s) \right] = (-p_s \bar{I} + \vec{\tau}_s) : \nabla \vec{v}_s + \nabla \cdot (k_{bs} \nabla \theta_s) - \gamma_{bs} + \mathcal{O}_{gs} \quad (7)$$

$$k_{bs} = \frac{15 d_s \rho_s \alpha_s \sqrt{\theta_s \pi}}{4(41 - 33\eta)} \left[ 1 + \frac{12}{5} \eta^2 (4\eta - 3) \alpha_s g_{0,ss} + \frac{16}{15\pi} \eta^2 (41 - 33\eta) \eta \alpha_s g_{0,ss} \right] \quad (8)$$

$$\eta = \frac{1}{2} (1 + e_{ss}) \gamma_{bs} = \frac{12(1 - e_{ss}^2)}{d_s \sqrt{\pi}} \rho_s \alpha_s^2 \theta_s^{3/2} \quad (9)$$


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Constitutive equations for closure of momentum conservation

$$\text{Solid pressure (Lun et al): } p_s = \alpha_s \rho_s \theta_s + 2\rho_s (1 + e_{ss}) \alpha_s^2 \theta_s g_{0,ss} \quad (10)$$

$$\text{Radial distribution function (Lun et al): } g_{0,ss} = \left[ 1 - \left( \frac{\alpha_s}{\alpha_{s,max}} \right)^3 \right]^{-1} \quad (11)$$

$$\text{Solid shear viscosity: } \mu_s = \mu_{s,col} + \mu_{s,kin} + \mu_{s,fr} \quad (12)$$

$$\text{Collisional viscosity (Gidaspow et al): } \mu_{s,col} = \frac{4}{5} \alpha_s \rho_s d_s g_{0,ss} (1 + e_{ss}) \sqrt{\frac{\theta_s}{\pi}} \quad (13)$$

$$\text{Kinetic viscosity (Syamlal et al): } \mu_{s,kin} = \frac{\alpha_s \rho_s d_s \sqrt{\theta_s \pi}}{6(3 + e_{ss})} \left[ 1 + \frac{2}{5} (1 + e_{ss}) (3e_{ss} - 1) \alpha_s g_{0,ss} \right] \quad (14)$$

$$\text{Frictional viscosity (Schaeffé): } \mu_{s,fr} = \frac{p_s \sin \phi}{2\sqrt{I_{2D}}} \quad (15)$$

$$\text{Bulk viscosity (Lun et al): } \lambda_s = \frac{4}{3} \alpha_s^2 \rho_s d_s g_{0,ss} (1 + e_{ss}) \sqrt{\frac{\theta_s}{\pi}} \quad (16)$$

Interphase momentum exchange coefficient (Gibilaro):

$$K_{gs} = \left( \frac{18}{Re} + 0.33 \right) \frac{\rho_g |v_s - v_g|}{d_p} \alpha_s \alpha_g^{-1.8} \quad (17)$$

$$Re = \frac{\alpha_g \rho_g d_p |v_s - v_g|}{\mu_g} \quad (18)$$


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capture system operates in the fast fluidization regime, such that high superficial velocity induces bypass of dry sorbent and leads to inefficiency of the system. Therefore, the present study seeks to increase the residence time of dry sorbent in the adsorption reactor of a multi-stage CO<sub>2</sub> capture system. First,

we conducted sensitivity analysis according to operating conditions and predicted the residence time of solid particles. Second, we applied the diffuser in the original reactor. Using CFD simulation, a comparison study was conducted to figure out the effect of the diffuser in the carbonation reactor of a

multi-stage CO<sub>2</sub> capture system.

## 2. Research method

An unsteady simulation was used to simulate gas–solid multiphase flow in the CFB riser using a Eulerian–Eulerian approach with kinetic theory of granular flow implemented using ANSYS Fluent version 14.0 [7–9]. A fixed time step of 10<sup>−3</sup> second was used. The model treats gas and solid phases as interpenetrating continua, which means that the presence of two phases is made possible in a single control volume via the introduction of fractional volume variables. Mass, momentum and species were conserved individually for each phase. Details of governing equations are listed in Table 1.

The solid phase was assumed to be a continuum, such that constitutive correlations were required for conservation of momentum; these were derived from the kinetic theory of granular flow. The granular temperature was introduced, which is proportional to the kinetic energy associated with the random motion of the particles. The pressure and viscosity of the solid were described as a function of the granular temperature to close conservation of momentum [7].

Forces due to drag acting on the particles in the gas–solid flow are represented by the final term on the right-hand side of the momentum conservation relation. It is composed of the product of the interphase momentum exchange coefficient,  $K_{gs}$ , and the slip velocity. The Gibilaro drag model was used, which has been shown to accurately describe the gas–solid flow characteristics in fluidized beds under turbulent flow conditions [10, 11].

## 3. Description of geometry and operating conditions

### 3.1 CFB riser for CO<sub>2</sub> capture

The multi-stage CO<sub>2</sub> capture system comprises three stages, low, mid, and high temperature. This study focuses on the bench-scale carbonation reactor of the mid-temperature stage [4]. Fig. 2 shows the carbonation reactor of a multi-stage CO<sub>2</sub> capture system and the directions of heat flows. The height of the bench-scale carbonation reactor is 6.8 m and the diameter is 7 cm. Two types of heat transfer take place in one reactor. In the bottom part of the reactor, reaction heat is transferred for the regeneration energy of used-sorbents of the lower stage. It has a double-pipe shape. The upper part transfers sensible heat from the regenerated sorbent at a high temperature to the carbonation reactor. It is a multi-pipe shape for increasing heat transfer surface. Geometric information on the bench-scale carbonation reactor and the physical properties of solid sorbents are summarized in Table 2.

### 3.2 Operating conditions

CFB risers typically operate at high superficial gas velocity, which results in entrainments of solid particles, which escape from the reactor [12, 13]. The carbonation reactor at the mid-

Table 2. Geometric parameters of CFB riser and properties of solid particles.

CFB riser for CO <sub>2</sub> capture	
Total height of carbonator	6.8 m
Diameter of bottom part [Single-pipe]	70.3 mm
Height of bottom part [Single-pipe]	4 m
Diameter of upper part [Multi-pipe]	23 mm
Height of bottom part [Multi-pipe]	2 m
Solid particles (Magnesium oxide)	
Diameter ( $d_p$ )	100 $\mu$ m
Density ( $\rho$ )	1606.66 kg/m <sup>3</sup>
Geldart classification	A group

Table 3. Boundary conditions for sensitivity analysis.

	Case 1	Case 2	Case 3
Sorbent circulation rate	350 kg/hr	350 kg/hr	350 kg/hr
Operating temperature	250°C	250°C	250°C
Superficial velocity	1.6 m/s	2.8 m/s	4.3 m/s
Fluidization regime	Turbulent	Fast	Pneumatic transport

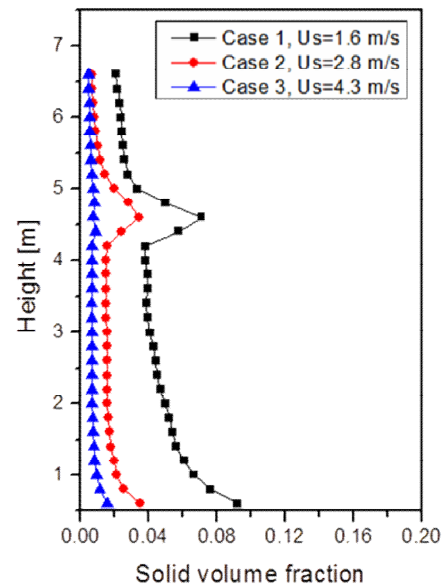


Fig. 2. CFB riser for CO<sub>2</sub> capture and directions of heat flow.

temperature stage operates at 250°C [5]. For sensitivity analysis for operating conditions, superficial gas velocities were changed. Boundary conditions for simulations are summarized in Table 3. Each superficial gas velocity belongs to the turbulent, fast fluidization regime, and pneumatic transport, respectively [14].

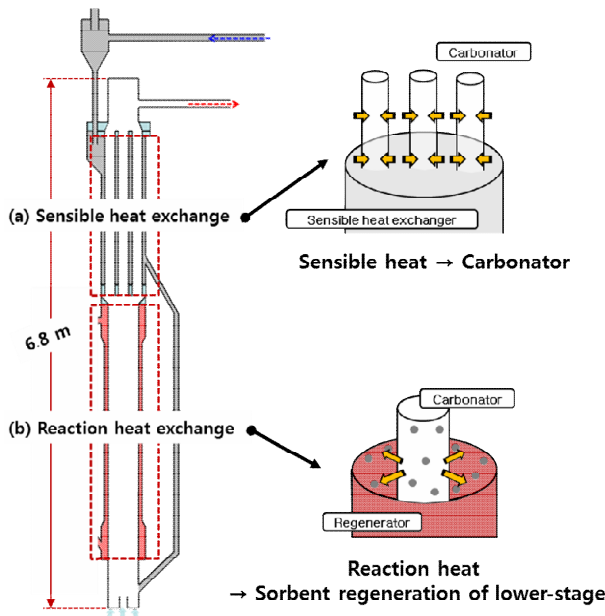


Fig. 3. Solid volume fraction according to operating conditions [Time-averaged].

4. Results

4.1 Solid flow characteristics according to operating conditions

Fig. 3 shows time-averaged solid volume fraction according to operating conditions. Generally, the amount of solid particles in the carbonation reactor was reduced with superficial gas velocity. The solid volume fraction in the cross-sectional area is presented in Fig. 4. The bottom part, the reaction heat exchanger, shows that more solid particles are located in the wall region because of the boundary layer. It shows a typical gas-solid flow characteristic in the CFB riser, called a core-annulus structure. Also, in the interconnecting region between the reaction and sensible heat exchanger, at 4.5 m, there is a transient increase in solid particles because of the deceleration of the gas phase. In the upper region, the amount of solid particles was reduced because the sensible heat exchanger is composed of multiple pipes and the cross-sectional area is reduced to that of the reaction heat exchanger.

As mentioned above, the CFB riser generally operates at high gas velocity. It has a unique flow structure, which is called the core-annulus structure. Because of the boundary layer in the wall region, particles are clustered, which results in a negative velocity in the wall region. Solid particles move upward at the center of the carbonation reactor because of drag force [15]. Therefore, this negative velocity increases the residence time of solid particles in the gas phase. Fig. 5 shows the profile of solid particles in the carbonation reaction. A denser wall region results in a larger area of the downward flow, which directly influences the residence time of solid particles. In the next chapter, comparison of the residence time of solid particles is discussed.

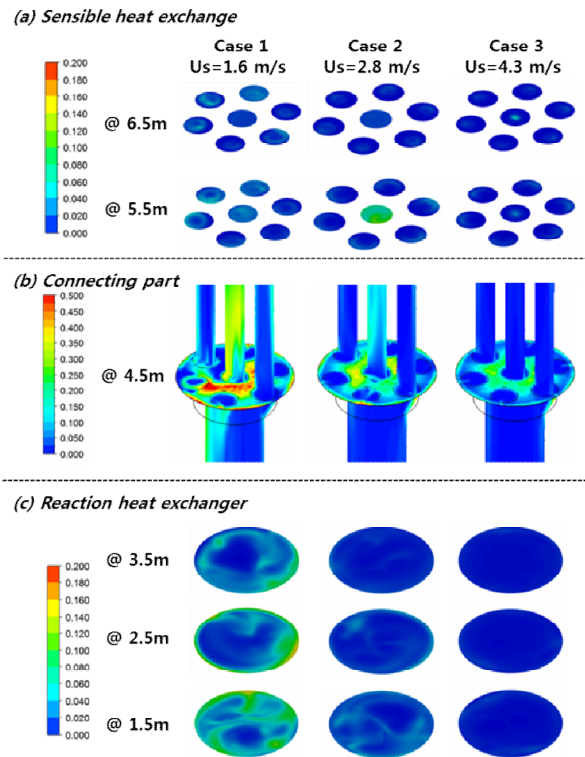


Fig. 4. Solid distribution according to operating conditions [Constant time at 60 sec].

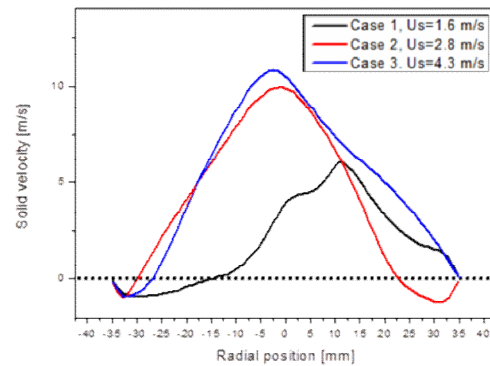


Fig. 5. Solid velocity at 2.5 m according to the operating conditions [Constant time at 60 sec].

4.2 Solid residence time according to operating conditions

As mentioned, for CO<sub>2</sub> capture reaction with dry sorbent, the residence time of solid particles is essential, and, in this chapter, we predict the residence time depending on the operating conditions as shown in Fig. 6.

The average residence time of solid particles can be estimated from the CFB riser volume occupied by solid particles divided by the volumetric flow of particles in the reactor [16].

$$t_{m,p} = \frac{\text{Reactor volume occupied by particles}}{\text{Volumetric flow of particles}} = \frac{AL[\rho_p(1-\bar{\epsilon})]}{AG_s} = \frac{\Delta P_{\text{riser}}}{G_s g} \quad (19)$$

Table 4. Boundary conditions and geometric parameters of the diffuser (2D simulation).

	Case 1	Case 2	Case 3
Diameter of diffuser	-	99.4 mm	99.4 mm
Length of diffuser	-	703 mm	703 mm
Angle of diffuser	-	6°	15°
Solid circulation rate	350 kg/hr	350 kg/hr	350 kg/hr
Superficial velocity	2.8 m/s	2.8 m/s	2.8 m/s

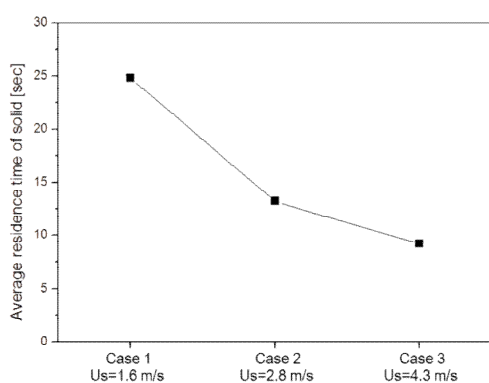


Fig. 6. Average residence time of solid particles in the carbonation reactor.

Average residence time of solid particles in the cases of turbulent, fast fluidization, and pneumatic transport are 24.8, 13.2, and 9.2 second, respectively. Increasing superficial gas velocity unavoidably decreases the residence time of particles. This means that, in order to handle more flue gas, a method of increasing the residence time of solid particles is necessary.

#### 4.3 Effect of the diffuser on solid flow in a CFB riser

The residence time of solid particles is an essential parameter in a fluidized bed reactor system. To increase the residence time of solid particles, the diffuser is applied in the carbonation reactor of a multi-stage CO<sub>2</sub> capture system. To check the performance and angle of the diffuser, a two-dimensional simulation was conducted. The center of the carbonation reactor was described and the same bed material, magnesium oxide sorbent, was used. The diffuser was applied at the center of the reaction heat exchanger in the carbonation reactor. The diffuser angle was changed. Information on the diffuser is summarized in Table 4.

First, Fig. 7 shows the time-averaged solid volume fraction. In the diffuser area, the solid volume fraction is increased, especially in the wall region. Case 3 with a larger diffuser angle has more solid volume fraction, which consequently means longer residence time in the reactor. For analysis of the diffuser angle, the right side of Fig. 7 shows a gas streamline at the inlet of the diffuser. Gas flow in cases 1 and 2 passes smoothly, but case 3 with the larger diffuser angle has separate flows at the inlet of the diffuser, such that the amount of

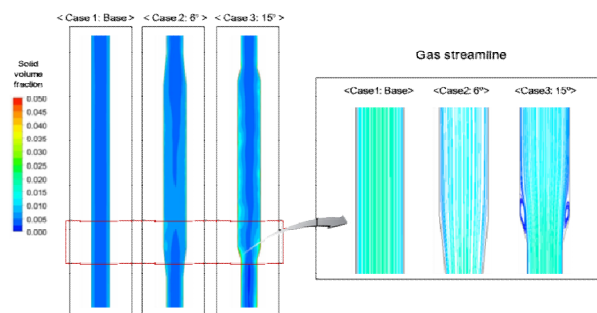


Fig. 7. Solid volume fraction and gas streamline in the diffuser according to the angle of the diffuser.

gas flow was decreased in the wall region. Therefore, case 3 results in more solid volume fraction in the wall region than cases 1 and 2. Applying the diffuser in the carbonation reactor increases the residence time of solid particles, as confirmed in this chapter. The diffuser angle affects the residence time considerably, as well.

## 5. Conclusion

A new CO<sub>2</sub> capture process, a multi-stage CO<sub>2</sub> capture system, was introduced to reduce the cost of CO<sub>2</sub> capture. This system's main concept is the use of the reaction heat of CO<sub>2</sub> capture as the regeneration energy for the lower stage. This study mainly focused on the residence time of solid particles in the bench-scale carbonation reactor of a multi-stage CO<sub>2</sub> capture system because of the important reaction characteristics of CO<sub>2</sub> capture.

First, sensitivity analysis depending on operating conditions was conducted to predict the average solid residence time in a bench-scale carbonation reactor, resulting in 24.8, 13.2, and 9.2 seconds in turbulent, fast fluidization and pneumatic transport, respectively. Also, a diffuser was applied to the carbonation reactor to increase the residence time of solid particles. The diffuser angle affected the residence time and the larger angle (15° with separation phenomena) was more effective at increasing the residence time.

Therefore, in this study, an effective way to increase the residence time of solid particles was proposed, which is expected to increase the amount of flue gas handled by the diffuser in the carbonation reactor.

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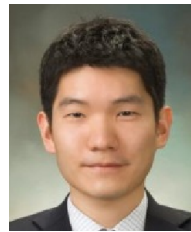
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## Nomenclature

$d_p$	: Particle diameter [m]
$g$	: Gravity acceleration [ $\text{m/s}^2$ ]
$g_0$	: Radial distribution coefficient
$G_s$	: Solid circulation rate [ $\text{kg} / \text{m}^2\text{s}$ ]
$p$	: Pressure [Pa]
$Re_s$	: Particle Reynolds number
$t$	: Time [s]
$\vec{v}$	: Velocity vector [m/s]
$U_s$	: Superficial gas velocity [m/s]
$\theta_s$	: Granular temperature
$\alpha$	: Volume fraction
$\rho$	: Density [ $\text{kg/m}^3$ ]
$\mu$	: Viscosity [Pa s]
$\tau_s$	: Stress tensor
$K_{gs}$	: Interchange momentum exchange coefficient

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